Supporting Information: Light-Matter Response in Non-Relativistic Quantum Electrodynamics

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October 29, 2019

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S1 Current state of the art for spectroscopy: semi-classical description

To highlight the many differences of the presented framework to the standard linear-response approach we give here a brief recapitulation of the standard (matter-only) theory. The current theoretical description of linear spectroscopic techniques is built on the *semi-classical* approximation.[S1] Herein, the many-particle electronic system is treated quantum mechanically while the nuclei are subject to the Born-Oppenheimer approximation and the electromagnetic field appears as an external perturbation. As an external perturbation, the electromagnetic field probes the quantum system, but is not a dynamical variable of the complete system. To arrive at the semi-classical description starting from the full non-relativistic description of the Pauli-Fierz Hamiltonian,[S2] several approximations are used to simplify the problem. In the following, we list these approximations explicitly

- The mean-field approximation renders the Pauli-Fierz Hamiltonian as a problem of two coupled equations, i.e. the time-dependent Pauli equation and the inhomogeneous Maxwell's equations, and is also know as the Maxwell-Pauli equation.[S3]
- The decoupling of these Maxwell-Pauli equations leads to the inhomogeneous Maxwell's equation becoming independent of the electronic system and all field effects are treated as a classical external field that perturbs the many-electron system.
- The dipole approximation, which ensures the uniformity of the external (decoupled) field over the extend of the electronic system.

Based on these approximations the Pauli-Fierz Hamiltonian[S3] reduces to the time-dependent semi-classical Hamiltonian for many-particle systems given as

$$\hat{H}_e(t) = \sum_{i=1}^{N}\left(-\frac{\hbar^2}{2m_e}\boldsymbol{\nabla}_i^2 + v(\mathbf{r}_i,t)\right) + \frac{e^2}{4\pi\epsilon_0}\sum_{i>j}^{N}\frac{1}{|\mathbf{r}_i-\mathbf{r}_j|}, \tag{S1}$$

including the kinetic energy, time-dependent external potential and the longitudinal Coulomb interaction. The time-dependent external potential has two parts $v(\mathbf{r},t) = v_0(\mathbf{r}) + \delta v(\mathbf{r},t)$. Here, $v_0(\mathbf{r})$ describes the attractive part of the external potential due to the nuclei and $\delta v(\mathbf{r},t) = e\mathbf{r}\cdot\mathbf{E}_\perp(t)$ with $\mathbf{E}_\perp(t)$ being a classical external (transversal) probe field in dipole approximation that couples to the electronic subsystem. In this decoupling limit of light and matter, the many-particle wavefunction is labeled only by the particle coordinate and spin as $\Psi(\mathbf{r}_1\sigma_1,...,\mathbf{r}_N\sigma_N)$. In the dipole approximation we can investigate dipole-related spectroscopic observables such as polarizabiltiy, absorption and emission spectra, etc from linear to all orders in the external perturbation. Consider the particular case of a response of an electronic system to an external weak probe field. In the dipole limit a key observable in the study of electronic and optical excitations in large many-particle systems is the electron density. Formulated within linear-response, the density response to an external perturbation is given as:[S4]

$$\begin{aligned}\delta n(\mathbf{r}t) &= -\frac{i}{\hbar}\int_{t_0}^{t}dt'\int d\mathbf{r}'\langle\Psi_0|\left[\hat{n}_I(\mathbf{r}t),\hat{n}_I(\mathbf{r}'t')\right]|\Psi_0\rangle \\ &= \int_{t_0}^{t}dt'\int d\mathbf{r}'\tilde{\chi}_n^n(\mathbf{r}t,\mathbf{r}'t')\delta v(\mathbf{r}'t').\end{aligned} \tag{S2}$$

Here, $\tilde{\chi}_n^n(\mathbf{r}t,\mathbf{r}'t')$ is the density-density function with respect to the ground-state $\Psi_0(\mathbf{r}_1\sigma_1,...,\mathbf{r}_N\sigma_N)$. Practical calculations for the response of a many-electron system is a considerable challenge due to the large degrees of freedom. In practice, time-dependent density functional theory (TDDFT)[S5,S6] is one of the most frequently applied theories to approach this problem. Knowing the electron density in TDDFT we can in principle calculate all observables of interest. Formulated within TDDFT linear-response, the density-density response function of the interacting system can be expressed in terms of non-interacting the density-density response function and an exchange-correlation (xc) kernel that has a form of a Dyson-type equation:[S7]

$$\tilde{\chi}_n^n(\mathbf{r}t,\mathbf{r}'t') = \chi_{n,\mathrm{s}}^n(\mathbf{r}t,\mathbf{r}'t') + \iint \mathrm{d}\mathbf{x}\mathrm{d}\tau \iint \mathrm{d}\tau'\mathrm{d}\mathbf{y}\chi_{n,\mathrm{s}}^n(\mathbf{r}t,\mathbf{x}\tau) f_{\mathrm{Hxc}}(\mathbf{x}\tau,\mathbf{y}\tau')\tilde{\chi}_n^n(\mathbf{y}\tau',\mathbf{r}'t'), \tag{S3}$$

where $\chi_{n,\mathrm{s}}^n$ and $f_{\mathrm{Hxc}} = \left(\chi_{n,\mathrm{s}}^n\right)^{-1} - (\tilde{\chi}_n^n)^{-1}$. One of the most widely employed approaches to TDDFT linear-response is the Casida formalism which can be written in a compact matrix form. The Casida equation obtains the exact excitation energies Ω_q of the many-particle system and requires all occupied and unoccupied Kohn-Sham orbitals and energies including the continuum of states. In practice, the Casida equation is often cast into the following form

$$U\mathbf{E} = \Omega_q^2\mathbf{E}. \tag{S4}$$

The explicit form of the matrix elements is given as (with $q = (i,a)$)

$$\begin{aligned} U_{qq'} &= \delta_{qq'}\omega_q^2 + 2\sqrt{\omega_q\omega_{q'}}K_{qq'}(\Omega_q), \\ K_{ai,jb}(\Omega_q) &= \iint d\mathbf{r}d\mathbf{r}'\varphi_i(\mathbf{r})\varphi_a^*(\mathbf{r})f_{Hxc}(\mathbf{r},\mathbf{r}',\Omega_q)\varphi_b(\mathbf{r}')\varphi_j^*(\mathbf{r}'). \end{aligned} \tag{S5}$$

The Casida formalism is well established and has been applied to a variety of systems, see e.g. Refs.[S8–S12] and references therein.

The many obvious shortcomings of the approximations that lead to the standard Schrödinger equation (S1) are well-known and discussed to some extend in the main part of the paper (for more details see, e.g., Ref.[S3]). We point out that all of the above ubiquitous fundamental equations are modified and the results based on the introduced generalized equations can differ strongly, as discussed in Sec. 3 of the main article.

S2 Linear-response in non-relativistic QED

To help the reader with the unfamiliar generalized linear-response framework for coupled light-matter systems, we here derive the linear-response equations and the ensuing response functions presented in Sec. 1. In the non-relativistic setting of QED, the static and dynamical behavior of the coupled electron-photon systems is given by

$$\hat{H}(t) = \hat{H}_0 + \hat{H}_{ext}(t). \tag{S6}$$

Where we define the time-independent electron-photon Hamiltonian as

$$\hat{H}_0 \;=\; \hat{T} + \hat{W}_{ee} + \frac{1}{2}\sum_{\alpha=1}^{M}\left[\hat{p}_\alpha^2 + \omega_\alpha^2\left(\hat{q}_\alpha - \frac{\boldsymbol{\lambda}_\alpha}{\omega_\alpha}\cdot\mathbf{R}\right)^2\right] + \sum_{i=1}^{N} v_0(\mathbf{r}_i) + \sum_{\alpha=1}^{M}\frac{j_{\alpha,0}}{\omega_\alpha}\hat{q}_\alpha, \tag{S7}$$

where the kinetic energy operator is $\hat{T} = -\frac{\hbar^2}{2m_e}\sum_{i=1}^{N}\boldsymbol{\nabla}_i^2$, 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 the time-dependent external perturbation is given by

$$\hat{H}_{ext}(t) = \hat{V}_{ext}(t) + \hat{J}_{ext}(t). \tag{S8}$$

Here, the time-dependent external potential and current are

$$\hat{V}_{ext}(t) = \sum_{i=1}^{N} v(\mathbf{r}_i, t), \quad \hat{J}_{ext}(t) = \sum_{\alpha}\frac{j_\alpha(t)}{\omega_\alpha}\hat{q}_\alpha. \tag{S9}$$

We now introduce the interaction picture, where a general state vector of the interacting electron-photon system is given by

$$\Psi_I(t) = \hat{U}_0^\dagger(t)\Psi(t) = e^{i\hat{H}_0 t/\hbar}\Psi(t),$$

with $\Psi(t)$ as the state vector in the Schrödinger picture. Accordingly, an arbitrary operator

$\hat{O}$ can be transformed from the Schrödinger to the interaction picture by

$$\hat{O}_I(t) = \hat{U}_0^\dagger(t)\hat{O}\hat{U}_0(t). \tag{S10}$$

In the interaction picture, the evolution of the interacting electron-photon system from an initial state Ψ_0 is described by the following time-dependent Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\Psi_I(t) = \hat{H}_{ext,I}(t)\Psi_I(t). \tag{S11}$$

Through an integration, the above equation can be formally solved to yield

$$\Psi_I(t) = \Psi_0 - \frac{i}{\hbar}\int_{t_0}^{t} dt'\hat{H}_{ext,I}(t')\Psi_I(t'). \tag{S12}$$

If we only keep the first order, we obtain in the Schrödinger picture a closed solution

$$\Psi(t) \simeq \hat{U}_0(t)\Psi_0 - \frac{i}{\hbar}\hat{U}_0(t)\int_{t_0}^{t} dt'\hat{H}_{ext,I}(t')\Psi_0. \tag{S13}$$

In our case however, we are not interested in the time evolution of the wave function, but rather in the response of an observable $\hat{O}$ to (small) external perturbations. The change in the expectation value of an arbitrary observable $\hat{O}$ due to the external perturbation $\hat{H}_{ext}(t)$ is given by

$$\delta\langle\hat{O}(t)\rangle = \langle\Psi(t)|\hat{O}|\Psi(t)\rangle - \langle\Psi_0|\hat{O}|\Psi_0\rangle, \tag{S14}$$

In linear-response theory, we now assume that the external perturbation in Eq. (S9) is sufficiently small such that Eq. (S13) is a good approximation to Eq. (S12) and that Ψ_0 equals the ground-state of Eq. (S7). Thus, if we evaluate Eq. (S14) with Eq. (S13), we obtain

$$\delta\langle\hat{O}(t)\rangle = -\frac{i}{\hbar}\int_{t_0}^{t} dt'\langle\Psi_0|\left[\hat{O}_I(t), \hat{H}_{ext,I}(t')\right]|\Psi_0\rangle, \tag{S15}$$

As a side remark, beyond linear-response solutions can be obtained by higher-order terms in

Eq. (S12). Staying within linear response, we can now use Eq. (S15) to obtain the response of the electron density to $\hat{H}_{ext}(t)$ that is given by

$$\delta n(\mathbf{r}t) = -\frac{i}{\hbar}\int_{t_0}^{t} dt' \int d\mathbf{r}' \langle\Psi_0| \left[\hat{n}_I(\mathbf{r}t), \hat{V}_{ext,I}(\mathbf{r}'t')\right] |\Psi_0\rangle - \frac{i}{\hbar}\sum_{\alpha}\int_{t_0}^{t} dt' \langle\Psi_0| \left[\hat{n}_I(\mathbf{r}t), \hat{J}_{ext,I}(t')\right] |\Psi_0\rangle.$$

Simplifying further, the density response reads

$$\begin{aligned}\delta n(\mathbf{r}t) &= -\tfrac{i}{\hbar}\textstyle\int_{t_0}^{t} dt' \int d\mathbf{r}' \langle\Psi_0| \left[\hat{n}_I(\mathbf{r}t), \hat{n}_I(\mathbf{r}'t')\right] |\Psi_0\rangle \delta v(\mathbf{r}'t') \\ &- \tfrac{i}{\hbar}\textstyle\sum_{\alpha}\int_{t_0}^{t} dt' \tfrac{1}{\omega_\alpha} \langle\Psi_0| \left[\hat{n}_I(\mathbf{r}t), \hat{q}_{\alpha,I}(t')\right] |\Psi_0\rangle \delta j_\alpha(t').\end{aligned}$$

The response of the density to the external perturbation $(v(\mathbf{r}t), j_\alpha(t))$ is

$$\delta n(\mathbf{r}t) = \int_{t_0}^{\infty} dt' \int d\mathbf{r}' \chi_n^n(\mathbf{r}t, \mathbf{r}'t') \delta v(\mathbf{r}'t') + \sum_{\alpha}\int_{t_0}^{\infty} dt' \chi_{q_\alpha}^n(\mathbf{r}t, t') \delta j_\alpha(t'),$$

where the response functions are

$$\chi_n^n(\mathbf{r}t, \mathbf{r}'t') = -\frac{i}{\hbar}\Theta(t-t')\langle\Psi_0| \left[\hat{n}_I(\mathbf{r}t), \hat{n}_I(\mathbf{r}'t')\right] |\Psi_0\rangle, \quad \text{(S16)}$$

$$\chi_{q_\alpha}^n(\mathbf{r}t, t') = -\frac{i}{\hbar}\Theta(t-t')\frac{1}{\omega_\alpha}\langle\Psi_0| \left[\hat{n}_I(\mathbf{r}t), \hat{q}_{\alpha,I}(t')\right] |\Psi_0\rangle. \quad \text{(S17)}$$

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

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

Following similar steps as above, the response of the photon coordinate to the external perturbation $(v(\mathbf{r}t), j_\alpha(t))$ is

$$\delta q_\alpha(t) = \int_{t_0}^{\infty} dt' \int d\mathbf{r}' \chi_n^{q_\alpha}(t, \mathbf{r}'t') \delta v(\mathbf{r}'t') + \sum_{\alpha'}\int_{t_0}^{\infty} dt' \chi_{q_{\alpha'}}^{q_\alpha}(t, t') \delta j_{\alpha'}(t'),$$

where the response functions are

$$\chi_n^{q_\alpha}(t,\mathbf{r}'t') = -\frac{i}{\hbar}\Theta(t-t')\langle\Psi_0|\,[q_{\alpha,I}(t),\hat{n}_I(\mathbf{r}'t')]\,|\Psi_0\rangle, \tag{S18}$$

$$\chi_{q_{\alpha'}}^{q_\alpha}(t,t') = -\frac{i}{\hbar}\Theta(t-t')\frac{1}{\omega_{\alpha'}}\langle\Psi_0|\,[q_{\alpha,I}(t),\hat{q}_{\alpha',I}(t')]\,|\Psi_0\rangle. \tag{S19}$$

Alternatively, the response functions of Eqs.(S16)-(S19) can be obtained using the functional dependence of the observables on the external pair $(v(\mathbf{r}t), j_\alpha(t))$. The wave function of Eq. (2) in the main manuscript has a functional dependence $\Psi([v,j_\alpha];t)$ via the Hamiltonian Eq. (S6), i.e., $\hat{H}(t) = \hat{H}([v,j_\alpha];t)$. Therefore, through the expectation of electron density and photon displacement coordinate, both have a functional dependence on the external pair as $n([v,j_\alpha];\mathbf{r}t)$ and $q_\alpha([v,j_\alpha];t)$, respectively.

Considering the ground-state problem with external potential and current of $(v_0(\mathbf{r}), j_{\alpha,0})$, we can perform a functional Taylor expansion of the density $n(\mathbf{r}t)$ and photon coordinate $q_\alpha(t)$ to first-order as

$$\begin{aligned}
n([v,j_\alpha];\mathbf{r}t) &= n([v_0,j_{\alpha,0}];\mathbf{r}) + \iint d\mathbf{r}'dt'\frac{\delta n([v_0,j_{\alpha,0}];\mathbf{r}t)}{\delta v(\mathbf{r}'t')}\delta v(\mathbf{r}'t') + \sum_\alpha \int dt'\frac{\delta n([v_0,j_{\alpha,0}];\mathbf{r}t)}{\delta j_\alpha(t')}\delta j_\alpha(t'),\\
q_\alpha([v,j_\alpha];t) &= q_\alpha([v_0,j_{\alpha,0}]) + \iint d\mathbf{r}'dt'\frac{\delta q_\alpha([v_0,j_{\alpha,0}];t)}{\delta v(\mathbf{r}'t')}\delta v(\mathbf{r}'t') + \sum_{\alpha'} \int dt'\frac{\delta q_\alpha([v_0,j_{\alpha,0}];t)}{\delta j_{\alpha'}(t')}\delta j_{\alpha'}(t').
\end{aligned}$$

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

$$\delta n([v,j_\alpha];\mathbf{r}t) = \iint d\mathbf{r}'dt'\chi_v^n(\mathbf{r}t,\mathbf{r}'t')\delta v(\mathbf{r}'t') + \sum_\alpha \int dt'\chi_{j_\alpha}^n(\mathbf{r}t,t')\delta j_\alpha(t'),$$

and

$$\delta q_\alpha([v,j_\alpha];t) = \iint d\mathbf{r}'dt'\chi_v^{q_\alpha}(t,\mathbf{r}'t')\delta v(\mathbf{r}'t') + \sum_{\alpha'} \int dt'\chi_{j_{\alpha'}}^{q_\alpha}(t,t')\delta j_{\alpha'}(t'),$$

where we define the response functions of the above relation as

$$
\begin{aligned}
\chi_v^n(\mathbf{r}t,\mathbf{r}'t') &= \left.\frac{\delta n([v,j_\alpha];\mathbf{r}t)}{\delta v(\mathbf{r}'t')}\right|_{v_0(\mathbf{r}),j_{\alpha,0}}, && \text{(S20)}\\
\chi_{j_\alpha}^n(\mathbf{r}t,t') &= \left.\frac{\delta n([v,j_\alpha];\mathbf{r}t)}{\delta j_\alpha(t')}\right|_{v_0(\mathbf{r}),j_{\alpha,0}}, && \text{(S21)}\\
\chi_v^{q_\alpha}(t,\mathbf{r}'t') &= \left.\frac{\delta q_\alpha([v,j_\alpha];t)}{\delta v(\mathbf{r}'t')}\right|_{v_0(\mathbf{r}),j_{\alpha,0}}, && \text{(S22)}\\
\chi_{j_{\alpha'}}^{q_\alpha}(t,t') &= \left.\frac{\delta q_\alpha([v,j_\alpha];t)}{\delta j_{\alpha'}(t')}\right|_{v_0(\mathbf{r}),j_{\alpha,0}}. && \text{(S23)}
\end{aligned}
$$

These response functions defined in Eqs.(S16)-(S19) and Eqs.(S20)-(S23) are equivalent.

The response functions expressed in the so-called Lehmann representation are given by

$$
\begin{aligned}
\chi_n^n(\mathbf{r},\mathbf{r}',\omega) &= \frac{1}{\hbar}\lim_{\eta\to0^+}\sum_k\left[\frac{f_k(\mathbf{r})f_k^*(\mathbf{r}')}{\omega-\Omega_k+i\eta}-\frac{f_k(\mathbf{r}')f_k^*(\mathbf{r})}{\omega+\Omega_k+i\eta}\right],\\
\chi_{q_\alpha}^n(\mathbf{r},\omega) &= \frac{1}{\hbar}\lim_{\eta\to0^+}\sum_k\frac{1}{\omega_\alpha}\left[\frac{f_k(\mathbf{r})g_{\alpha,k}^*}{\omega-\Omega_k+i\eta}-\frac{g_{\alpha,k}f_k^*(\mathbf{r})}{\omega+\Omega_k+i\eta}\right],\\
\chi_n^{q_\alpha}(\mathbf{r}',\omega) &= \frac{1}{\hbar}\lim_{\eta\to0^+}\sum_k\left[\frac{g_{\alpha,k}f_k^*(\mathbf{r}')}{\omega-\Omega_k+i\eta}-\frac{f_k(\mathbf{r}')g_{\alpha,k}^*}{\omega+\Omega_k+i\eta}\right],\\
\chi_{q_{\alpha'}}^{q_\alpha}(\omega) &= \frac{1}{\hbar}\lim_{\eta\to0^+}\sum_k\frac{1}{\omega_{\alpha'}}\left[\frac{g_{\alpha,k}g_{\alpha',k}^*}{\omega-\Omega_k+i\eta}-\frac{g_{\alpha',k}g_{\alpha,k}^*}{\omega+\Omega_k+i\eta}\right],
\end{aligned}
$$

where $f_k(\mathbf{r}) = \langle\Psi_0|\hat{n}(\mathbf{r})|\Psi_k\rangle$ 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. As a side remark, if we can choose the wave functions Ψ_0 and Ψ_k to be real, we find $g_{\alpha,k} = g_{\alpha,k}^*$, and $f_k(\mathbf{r}) = f_k^*(\mathbf{r})$, thus $\chi_n^{q_\alpha}(\mathbf{r},\omega) = \omega_\alpha\chi_{q_\alpha}^n(\mathbf{r},\omega)$.

S3 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 $(v(\mathbf{r}t), j_\alpha(t))$ are evaluated at the ground-state $(v_0(\mathbf{r}), j_{\alpha,0})$ and will not be written explicitly.

The non-interacting subsystems moving in an effective potential and current $(v_s(\mathbf{r}t), j^s_\alpha(t))$ can be written as a time-dependent problem of the Schrödinger

$$i\hbar\frac{\partial}{\partial t}\Phi(t) = \hat{H}_{\text{KS}}(t)\Phi(t). \tag{S24}$$

Here, $\Phi(t)$ is the wave function of the auxiliary non-interacting system and the non-interacting effective Hamiltonian $\hat{H}_{\text{KS}}(t) = \hat{H}^{(0)}_{\text{KS}} + \hat{H}^{(ext)}_{\text{KS}}(t)$ that is meant to reproduce the exact density and displacement field, is given explicitly as

$$\hat{H}^{(0)}_{\text{KS}} = \hat{T} + \hat{H}_{pt} + \left(v_0(\mathbf{r}) + v^{(0)}_{Mxc}([n, q_\alpha]; \mathbf{r})\right) + \sum_\alpha \frac{1}{\omega_\alpha}\left(j_{\alpha,0} + j^{(0)}_{\alpha,Mxc}[n, q_\alpha]\right)\hat{q}_\alpha,$$

and

$$\hat{H}^{(ext)}_{\text{KS}}(t) = (v(\mathbf{r}t) + v_{Mxc}([n, q_\alpha]; \mathbf{r}t)) + \sum_\alpha \frac{1}{\omega_\alpha}\left(j_\alpha(t) + j_{\alpha,Mxc}\left([n, q_\alpha]; t\right)\right)\hat{q}_\alpha.$$

Here $\hat{H}_{pt} = \frac{1}{2}\sum_{\alpha=1}^{M}[\hat{p}^2_\alpha + \omega^2_\alpha\hat{q}^2_\alpha]$ is the oscillator for the photon mode and the mean-field xc potential and current are defined as

$$v_{Mxc}([n, q_\alpha]; \mathbf{r}t) := v_s([n]; \mathbf{r}t) - v([n, q_\alpha]; \mathbf{r}t), \tag{S25}$$

$$j_{\alpha,Mxc}([n, q_\alpha]; t) := j^s_\alpha([q_\alpha]; t) - j_\alpha([n, q_\alpha]; t). \tag{S26}$$

In the above definitions of $v_{Mxc}([n, q_\alpha]; \mathbf{r}t)$ and $j_{\alpha,Mxc}([n, q_\alpha]; t)$, the initial state dependence of the interacting Ψ_0 and non-interacting Φ_0 system has been dropped. For completeness, the definition of $j_{\alpha,Mxc}([n, q_\alpha]; t)$ accounts for a functional dependence on q_α but this term can be calculated explicitly since it has no xc part as seen in Eq. (8) of the main manuscript.

The simplified form of $j_{\alpha,Mxc}$ is shown in Eq. (6) of the main manuscript.

Through similar steps as in Eqs.(S11)-(S13), in first-order the solution of the Schrödinger-Kohn-Sham equation reads

$$\Phi(t) \simeq \hat{U}_{\text{KS},0}(t)\Phi_0 - \frac{i}{\hbar}\hat{U}_{\text{KS},0}(t)\int_{t_0}^{t} dt' \hat{H}^{(ext)}_{\text{KS},I}(t')\hat{U}^{\dagger}_{\text{KS},0}(t)\Phi_0. \tag{S27}$$

where $\hat{U}_{\text{KS},0} = e^{-i\hat{H}^{(0)}_{\text{KS}}t/\hbar}$. Next, the bijective mapping between the interacting and non-interacting system that yields the same density and photon coordinate is given as

$$(v(\mathbf{r}t), j_\alpha(t)) \underset{\Psi_0}{\overset{1:1}{\longleftrightarrow}} (n(\mathbf{r}t), q_\alpha(t)) \underset{\Phi_0}{\overset{1:1}{\longleftrightarrow}} (v_s(\mathbf{r}t), j^s_\alpha(t)), \tag{S28}$$

which can be inverted as $(v_s([v, j_\alpha]; \mathbf{r}'t'), j^s_\alpha([v, j_\alpha]; t'))$. The response of the electronic subsystem due to the perturbations with the external pair $(v(\mathbf{r}t), j_\alpha(t))$ is

$$\begin{aligned}\delta n(\mathbf{r}t) &= -\frac{i}{\hbar}\iint d\tau d\mathbf{x} \iint dt' d\mathbf{r}' \langle\Phi_0| \left[\hat{n}_I(\mathbf{r}t), \hat{n}_I(\mathbf{x}\tau)\right] |\Phi_0\rangle \frac{\delta v_s([v, j_\alpha]; \mathbf{x}\tau)}{\delta v(\mathbf{r}'t')}\delta v(\mathbf{r}'t') \\ &\quad -\frac{i}{\hbar}\iint d\tau d\mathbf{x} \sum_\alpha \int dt' \langle\Phi_0| \left[\hat{n}_I(\mathbf{r}t), \hat{n}_I(\mathbf{x}\tau)\right] |\Phi_0\rangle \frac{\delta v_s([v, j_\alpha]; \mathbf{x}\tau)}{\delta j_\alpha(t')}\delta j_\alpha(t').\end{aligned}$$

Where $\langle\Phi_0| \left[\hat{n}_I(\mathbf{r}t), \hat{q}_{\alpha,I}(\tau)\right] |\Phi_0\rangle = 0$ since both, electronic and photonic subsystems, are independent in the non-interacting system. From Eq. (S28), we have $(v_s([n]; \mathbf{r}t), j^s_\alpha([q_\alpha]; t))$ such that the above equation becomes

$$\begin{aligned}\delta n(\mathbf{r}t) = &\iint d\tau d\mathbf{x} \iint dt' d\mathbf{r}' \iint d\tau' d\mathbf{y} \chi^n_{n,s}(\mathbf{r}t, \mathbf{x}\tau) \frac{\delta v_s([n]; \mathbf{x}\tau)}{\delta n(\mathbf{y}\tau')} \frac{\delta n([v, j_\alpha]; \mathbf{y}\tau')}{\delta v(\mathbf{r}'t')}\delta v(\mathbf{r}'t') \\ &+ \iint d\tau d\mathbf{x} \sum_\alpha \int dt' \iint d\tau' d\mathbf{y} \chi^n_{n,s}(\mathbf{r}t, \mathbf{x}\tau) \frac{\delta v_s([n]; \mathbf{x}\tau)}{\delta n(\mathbf{y}\tau')} \frac{\delta n([v, j_\alpha]; \mathbf{y}\tau')}{\delta j_\alpha(t')}\delta j_\alpha(t'),\end{aligned} \tag{S29}$$

where $\chi^n_{n,s}(\mathbf{r}t, \mathbf{x}\tau) = (-i/\hbar)\Theta(t - \tau)\langle\Phi_0| \left[\hat{n}_I(\mathbf{r}t), \hat{n}_I(\mathbf{x}\tau)\right] |\Phi_0\rangle$ is the non-interacting density-density response function. For clarity, the above density response is $\delta n(\mathbf{r}t) = \delta n_v(\mathbf{r}t) + \delta n_j(\mathbf{r}t)$, where $(\delta n_v(\mathbf{r}t), \delta n_j(\mathbf{r}t))$ is the density response to the external pair $(v(\mathbf{r}t), j_\alpha(t))$, respectively.

Using Eqs.(S25) and (S26), we define the mean-field xc kernels as:

$$f^{n}_{Mxc}([n,q_\alpha];\mathbf{r}t,\mathbf{r}'t') = \frac{\delta v_s([n];\mathbf{r}t)}{\delta n(\mathbf{r}'t')} - \frac{\delta v([n,q_\alpha];\mathbf{r}t)}{\delta n(\mathbf{r}'t')}, \tag{S30}$$

$$f^{q_\alpha}_{Mxc}([n,q_\alpha];\mathbf{r}t,t') = -\frac{\delta v([n,q_\alpha];\mathbf{r}t)}{\delta q_\alpha(t')}, \tag{S31}$$

$$g^{n}_{Mxc}([n,q_\alpha];t,\mathbf{r}'t') = -\frac{\delta j_\alpha([n,q_\alpha];t)}{\delta n(\mathbf{r}'t')}, \tag{S32}$$

$$g^{q_{\alpha'}}_{Mxc}([n,q_\alpha];t,t') = \frac{\delta j^s_\alpha([q_\alpha];t)}{\delta q_{\alpha'}(t')} - \frac{\delta j_\alpha([n,q_\alpha];t)}{\delta q_{\alpha'}(t')}, \tag{S33}$$

where $\frac{\delta v_s([n];\mathbf{r}t)}{\delta q_\alpha(t')} = 0 = \frac{\delta j^s_\alpha([q_\alpha];t)}{\delta n(\mathbf{r}'t')}$. These kernels are the respective inverse of the interacting and non-interacting response functions.

From Eq. (S29), density response to $\delta v(\mathbf{r}t)$ can be written in terms of the density-density response function given by

$$\begin{aligned}\chi^n_n(\mathbf{r}t,\mathbf{r}'t') &= \iint d\tau d\mathbf{x}\chi^n_{n,s}(\mathbf{r}t,\mathbf{x}\tau)\iint d\tau' d\mathbf{y} f^n_{Mxc}([n,q_\alpha];\mathbf{x}\tau,\mathbf{y}\tau')\frac{\delta n([v,j_\alpha];\mathbf{y}\tau')}{\delta v(\mathbf{r}t')} \\ &\quad + \iint d\tau d\mathbf{x}\chi^n_{n,s}(\mathbf{r}t,\mathbf{x}\tau)\iint d\tau' d\mathbf{y}\frac{\delta v([n,q_\alpha];\mathbf{x}\tau)}{\delta n(\mathbf{y}\tau')}\frac{\delta n([v,j_\alpha];\mathbf{y}\tau')}{\delta v(\mathbf{r}'t')}.\end{aligned}$$

Making the following substitution in the above equation

$$\iint d\mathbf{y}d\tau' \frac{\delta v([n,q_\alpha];\mathbf{x}\tau)}{\delta n(\mathbf{y}\tau')}\frac{\delta n([v,j_\alpha];\mathbf{y}\tau')}{\delta v(\mathbf{r}'t')} = \delta(\mathbf{x}-\mathbf{r}')\delta(\tau-t') - \sum_\alpha \int d\tau' \frac{\delta v([n,q_\alpha];\mathbf{x}\tau)}{\delta q_\alpha(\tau')}\frac{\delta q_\alpha([v,j_\alpha];\tau')}{\delta v(\mathbf{r}'t')},$$

where $\delta v([n,q_\alpha];\mathbf{x}\tau)/\delta v(\mathbf{r}'t') = \delta(\mathbf{x}-\mathbf{r}')\delta(\tau-t')$, we obtain the relation

$$\begin{aligned}\chi^n_n(\mathbf{r}t,\mathbf{r}'t') &= \chi^n_{n,s}(\mathbf{r}t,\mathbf{r}'t') + \iiiint d\tau d\mathbf{x} d\tau' d\mathbf{y}\chi^n_{n,s}(\mathbf{r}t,\mathbf{x}\tau) f^n_{Mxc}(\mathbf{x}\tau,\mathbf{y}\tau')\chi^n_n(\mathbf{y}\tau',\mathbf{r}'t') \\ &\quad + \sum_\alpha \iiint d\tau d\mathbf{x} d\tau' \chi^n_{n,s}(\mathbf{r}t,\mathbf{x}\tau) f^{q_\alpha}_{Mxc}(\mathbf{x}\tau,\tau')\chi^{q_\alpha}_n(\tau',\mathbf{r}'t').\end{aligned} \tag{S34}$$

Next, the density response to $\delta j_\alpha(t)$ in Eq. (S29) is expressed in terms of the response function as

$$\chi^n_{q_\alpha}(\mathbf{r}t,t') = \iint d\tau d\mathbf{x}\chi^n_{n,s}(\mathbf{r}t,\mathbf{x}\tau)\iint d\tau' d\mathbf{y} f^n_{Mxc}(\mathbf{x}\tau,\mathbf{y}\tau')\frac{\delta n([v,j_\alpha];\mathbf{y}\tau')}{\delta j_\alpha(t')} + \iint d\tau d\mathbf{x}\chi^n_{n,s}(\mathbf{r}t,\mathbf{x}\tau)\iint d\tau' d\mathbf{y}\frac{\delta v([n,q_\alpha];\mathbf{x}\tau)}{\delta n(\mathbf{y}\tau')}\frac{\delta n([v,j_\alpha];\mathbf{y}\tau')}{\delta j_\alpha(t')}.$$

Using the relation (obtained from $\delta v([n,q_\alpha];\mathbf{x}\tau)/\delta j_\alpha(t')$)

$$\iint d\mathbf{y}d\tau'\frac{\delta v([n,q_\alpha];\mathbf{x}\tau)}{\delta n(\mathbf{y}\tau')}\frac{\delta n([v,j_\alpha];\mathbf{y}\tau')}{\delta j_\alpha(t')} = -\sum_{\alpha'}\int d\tau'\frac{\delta v([n,q_\alpha];\mathbf{x}\tau)}{\delta q_{\alpha'}(\tau')}\frac{\delta q_{\alpha'}([v,j_\alpha];\tau')}{\delta j_\alpha(t')},$$

the response function is given as

$$\chi^n_{q_\alpha}(\mathbf{r}t,t') = \iiiint d\tau d\mathbf{x}d\tau' d\mathbf{y}\chi^n_{n,s}(\mathbf{r}t,\mathbf{x}\tau)f^n_{Mxc}(\mathbf{x}\tau,\mathbf{y}\tau')\chi^n_{q_\alpha}(\mathbf{y}\tau',t') + \sum_{\alpha'}\iiint d\tau d\mathbf{x}d\tau'\chi^n_{n,s}(\mathbf{r}t,\mathbf{x}\tau)f^{q_{\alpha'}}_{Mxc}(\mathbf{x}\tau,\tau')\chi^{q_{\alpha'}}_{q_\alpha}(\tau',t'). \tag{S35}$$

Similarly, the response to the photonic subsystem to linear perturbations from the external pair $(v(\mathbf{r}t), j_\alpha(t))$ is

$$\delta q_\alpha(t) = -\frac{i}{\hbar}\sum_\beta\int_{t_0}^t d\tau\frac{1}{\omega_\beta}\langle\Phi_0|\left[q_{\alpha,I}(t),q_{\beta,I}(\tau)\right]|\Phi_0\rangle\iint dt' d\mathbf{r}'\frac{\delta j^s_\beta([v,j_\alpha];\tau)}{\delta v(\mathbf{r}'t')}\delta v(\mathbf{r}'t') -\frac{i}{\hbar}\sum_\beta\int_{t_0}^t d\tau\frac{1}{\omega_\beta}\langle\Phi_0|\left[q_{\alpha,I}(t),q_{\beta,I}(\tau)\right]|\Phi_0\rangle\sum_{\alpha'}\int dt'\frac{\delta j^s_\beta([v,j_\alpha];\tau)}{\delta j_{\alpha'}(t')}\delta j_{\alpha'}(t'),$$

where $\langle\Phi_0|\left[\hat{q}_{\alpha,I}(t),\hat{n}_I(\mathbf{x}\tau)\right]|\Phi_0\rangle = 0$ in the non-interacting system. By defining the non-interacting photon-photon response function as $\chi^{q_\alpha}_{q_{\beta,s}}(t,\tau) = (-i/\hbar)\Theta(t-\tau)(1/\omega_\beta)\langle\Phi_0|\left[q_{\alpha,I}(t),q_{\beta,I}(\tau)\right]|\Phi_0\rangle$ and using Eq. (S28), where we have $(v_s([n];\mathbf{r}t), j^s_\alpha([q_\alpha];t))$, the response can be written as

$$\delta q_\alpha(t) = \sum_\beta\int d\tau\chi^{q_\alpha}_{q_{\beta,s}}(t,\tau)\sum_{\beta'}\iiint dt' d\mathbf{r}' d\tau'\frac{\delta j^s_\beta([q_\alpha];\tau)}{\delta q_{\beta'}(\tau')}\frac{\delta q_{\beta'}([v,j_\alpha];\tau')}{\delta v(\mathbf{r}'t')}\delta v(\mathbf{r}'t') + \sum_\beta\int d\tau\chi^{q_\alpha}_{q_{\beta,s}}(t,\tau)\sum_{\alpha',\beta'}\iint dt' d\tau'\frac{\delta j^s_\beta([q_\alpha];\tau)}{\delta q_{\beta'}(\tau')}\frac{\delta q_{\beta'}([v,j_\alpha];\tau')}{\delta j_{\alpha'}(t')}\delta j_{\alpha'}(t'). \tag{S36}$$

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

From Eq. (S36), the field response to $\delta v(\mathbf{r}t)$ can be written in terms of the photon-density response function as

$$\chi_n^{q_\alpha}(t,\mathbf{r}'t') = \sum_\beta \int d\tau \chi^{q_\alpha}_{q_{\beta,s}}(t,\tau) \sum_{\beta'} \int d\tau' g^{q_{\beta'}}_{Mxc}(\tau,\tau')\chi_n^{q_{\beta'}}(\tau',\mathbf{r}'t') + \sum_\beta \int d\tau \chi^{q_\alpha}_{q_{\beta,s}}(t,\tau) \sum_{\beta'} \int d\tau' \frac{\delta j_\beta([n,q_\alpha];\tau)}{\delta q_{\beta'}(\tau')} \frac{\delta q_{\beta'}([v,j_\alpha];\tau')}{\delta v(\mathbf{r}'t')}.$$

Using the relation (obtained from $\delta j_\beta([n,q_\alpha];\tau)/\delta v(\mathbf{r}'t')$)

$$\sum_{\beta'} \int d\tau' \frac{\delta j_\beta([n,q_\alpha];\tau)}{\delta q_{\beta'}(\tau')} \frac{\delta q_{\beta'}([v,j_\alpha];\tau')}{\delta v(\mathbf{r}t')} = -\iint d\tau' d\mathbf{y} \frac{\delta j_\beta([n,q_\alpha];\tau)}{\delta n(\mathbf{y}\tau')} \frac{\delta n([v,j_\alpha];\mathbf{y}\tau')}{\delta v(\mathbf{r}'t')},$$

the response function is given as

$$\chi_n^{q_\alpha}(t,\mathbf{r}'t') = \sum_\beta \int d\tau \iint d\tau' d\mathbf{y} \chi^{q_\alpha}_{q_{\beta,s}}(t,\tau) g^{n_\beta}_{Mxc}(\tau,\mathbf{y}\tau') \chi_n^n(\mathbf{y}\tau',\mathbf{r}'t'), \tag{S37}$$

where $g^{n_\beta}_{Mxc} = g^{n_\beta}_M$ and $g^{q_\alpha}_{Mxc} = 0$ as determined from the equation of motion for the displacement field. Also, from Eq. (S36), field response to δj_α can be written in terms of the photon-photon response function as

$$\chi^{q_\alpha}_{q_{\alpha'}}(t,t') = \sum_\beta \int d\tau \chi^{q_\alpha}_{q_{\beta,s}}(t,\tau) \sum_{\beta'} \int d\tau' g^{q_{\beta'}}_{Mxc}(\tau,\tau') \chi^{q_{\beta'}}_{q_{\alpha'}}(\tau',t') + \sum_\beta \int d\tau \chi^{q_\alpha}_{q_{\beta,s}}(t,\tau) \sum_{\beta'} \int d\tau' \frac{\delta j_\beta([n,q_\alpha];\tau)}{\delta q_{\beta'}(\tau')} \frac{\delta q_{\beta'}([n,q_\alpha];\tau')}{\delta j_{\alpha'}(t')}.$$

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

$$\sum_{\beta'} \int d\tau' \frac{\delta j_\beta([n,q_\alpha];\tau)}{\delta q_{\beta'}(\tau')} \frac{\delta q_{\beta'}([v,j_\alpha];\tau')}{\delta j_{\alpha'}(t')} = \delta(\tau - t')\delta_{\beta,\alpha'} - \iint d\tau' d\mathbf{x} \frac{\delta j_\beta([n,q_\alpha];\tau)}{\delta n(\mathbf{x}\tau')} \frac{\delta n([v,j_\alpha];\mathbf{x}\tau')}{\delta j_{\alpha'}(t')},$$

yields the photon-photon response function

$$\chi^{q_\alpha}_{q_{\alpha'}}(t,t') = \chi^{q_\alpha}_{q_{\alpha'},s}(t,t') + \sum_{\beta} \iiint d\tau d\tau' d\mathbf{x} \chi^{q_\alpha}_{q_\beta,s}(t,\tau) g^{n_\beta}_{Mxc}(\tau, \mathbf{x}\tau') \chi^{n}_{q_{\alpha'}}(\mathbf{x}\tau', t'), \tag{S38}$$

where $g^{q_{\beta'}}_{Mxc} = 0$ since $j_{\alpha,M}$ in Eq. (6) of the main manuscript has no functional dependency on q_α.

S4 Matrix formulation of QEDFT response equations

In this section we present a matrix formulation of non-relativistic QEDFT response equations which in the no-coupling limit reduces to Casida equation. Through a Fourier transform of Eqs.(S34)-(S35) and Eqs.(S37)-(S38) and making a substitution into Eqs.(35)-(38) (main manuscript), we express the responses in the following form:

$$\delta n_v(\mathbf{r},\omega) = \sum_{i,a} \left[\varphi_a(\mathbf{r})\varphi_i^*(\mathbf{r}) \mathbf{P}^{(1)}_{ai,v}(\omega) + \varphi_i(\mathbf{r})\varphi_a^*(\mathbf{r}) \mathbf{P}^{(1)}_{ia,v}(\omega) \right], \tag{S39}$$

$$\delta n_j(\mathbf{r},\omega) = \sum_{i,a} \left[\varphi_a(\mathbf{r})\varphi_i^*(\mathbf{r}) \mathbf{P}^{(1)}_{ai,j}(\omega) + \varphi_i(\mathbf{r})\varphi_a^*(\mathbf{r}) \mathbf{P}^{(1)}_{ia,j}(\omega) \right], \tag{S40}$$

$$\delta q_{\alpha,v}(\omega) = \mathbf{L}^{(1)}_{\alpha,v,-}(\omega) + \mathbf{L}^{(1)}_{\alpha,v,+}(\omega), \tag{S41}$$

$$\delta q_{\alpha,j}(\omega) = \mathbf{L}^{(1)}_{\alpha,j,-}(\omega) + \mathbf{L}^{(1)}_{\alpha,j,+}(\omega). \tag{S42}$$

Here, the subscripts (v,j) on the first-order responses $\mathbf{P}^{(1)}_{ia,v}$, $\mathbf{P}^{(1)}_{ia,j}$, $\mathbf{P}^{(1)}_{ai,v}$, $\mathbf{P}^{(1)}_{ai,j}$, $\mathbf{L}^{(1)}_{\alpha,v,\pm}$ and $\mathbf{L}^{(1)}_{\alpha,j,\pm}$ shows to what external perturbations $(\delta v(\mathbf{r},t), \delta j_\alpha(t))$ is being considered to induce the coupled responses. In defining Eqs.(S39)-(S42), we used the static KS orbitals in the Lehmann spectral representation of $\chi^n_{n,s}(\mathbf{r},\mathbf{r}',\omega)$ and photon-photon response function

$\chi^{q_\alpha}_{q_{\alpha,s}}(\omega)$ for a single-photon in Fock number basis are given as

$$\chi^{n}_{n,s}(\mathbf{r},\mathbf{r}',\omega) = \sum_{i,a}\left(\frac{\psi_a(\mathbf{r})\psi_i(\mathbf{r}')\psi_i^*(\mathbf{r})\psi_a^*(\mathbf{r}')}{\omega-(\epsilon_a-\epsilon_i)+i\eta} - \frac{\psi_i(\mathbf{r})\psi_a(\mathbf{r}')\psi_a^*(\mathbf{r})\psi_i^*(\mathbf{r}')}{\omega+(\epsilon_a-\epsilon_i)+i\eta}\right),$$
$$\chi^{q_\alpha}_{q_{\alpha,s}}(\omega) = \frac{1}{2\omega_\alpha^2}\left(\frac{1}{\omega-\omega_\alpha+i\eta} - \frac{1}{\omega+\omega_\alpha+i\eta}\right).$$

where the summations over occupied and unoccupied Kohn-Sham orbitals are performed according to $\sum_i = \sum_{i=1}^{N}$ and $\sum_a = \sum_{a=N+1}^{\infty}$ and from here on $\lim_{\eta\to 0^+}$ is implied. The first-order responses $\mathbf{P}^{(1)}_{ia,v}$, $\mathbf{P}^{(1)}_{ia,j}$, $\mathbf{P}^{(1)}_{ai,v}$, $\mathbf{P}^{(1)}_{ai,j}$, $\mathbf{L}^{(1)}_{\alpha,v,\pm}$ and $\mathbf{L}^{(1)}_{\alpha,j,\pm}$ are given by

$$[\omega-\omega_{ai}]\,\mathbf{P}^{(1)}_{ai,v}(\omega) = \int d\mathbf{r}\varphi_i(\mathbf{r})\varphi_a^*(\mathbf{r})\delta v^{(1)}_{\mathrm{KS},v}(\mathbf{r},\omega), \tag{S43}$$

$$[\omega+\omega_{ai}]\,\mathbf{P}^{(1)}_{ia,v}(\omega) = -\int d\mathbf{r}\varphi_a(\mathbf{r})\varphi_i^*(\mathbf{r})\delta v^{(1)}_{\mathrm{KS},v}(\mathbf{r},\omega), \tag{S44}$$

$$[\omega-\omega_{ai}]\,\mathbf{P}^{(1)}_{ai,j}(\omega) = \int d\mathbf{r}\varphi_i(\mathbf{r})\varphi_a^*(\mathbf{r})\delta v^{(1)}_{\mathrm{KS},j}(\mathbf{r},\omega), \tag{S45}$$

$$[\omega+\omega_{ai}]\,\mathbf{P}^{(1)}_{ia,j}(\omega) = -\int d\mathbf{r}\varphi_a(\mathbf{r})\varphi_i^*(\mathbf{r})\delta v^{(1)}_{\mathrm{KS},j}(\mathbf{r},\omega), \tag{S46}$$

$$[\omega-\omega_\alpha]\,\mathbf{L}^{(1)}_{\alpha,v,-}(\omega) = \frac{1}{2\omega_\alpha^2}\delta j^{(1)}_{\alpha,\mathrm{KS},v}(\omega), \tag{S47}$$

$$[\omega+\omega_\alpha]\,\mathbf{L}^{(1)}_{\alpha,v,+}(\omega) = -\frac{1}{2\omega_\alpha^2}\delta j^{(1)}_{\alpha,\mathrm{KS},v}(\omega), \tag{S48}$$

$$[\omega-\omega_\alpha]\,\mathbf{L}^{(1)}_{\alpha,j,-}(\omega) = \frac{1}{2\omega_\alpha^2}\delta j^{(1)}_{\alpha,\mathrm{KS},j}(\omega), \tag{S49}$$

$$[\omega+\omega_\alpha]\,\mathbf{L}^{(1)}_{\alpha,j,+}(\omega) = -\frac{1}{2\omega_\alpha^2}\delta j^{(1)}_{\alpha,\mathrm{KS},j}(\omega), \tag{S50}$$

where $\omega_{ai} = (\epsilon_a - \epsilon_i)$ and the respective effective potentials and currents $(\delta v_{s,\nu}(\mathbf{r},\omega), j^s_{\alpha,\nu}(\omega))$ as

$$\delta v^{(1)}_{\mathrm{KS},v}(\mathbf{r},\omega) = \delta v(\mathbf{r},\omega) + \int d\mathbf{r}' f^{n}_{Mxc}(\mathbf{r},\mathbf{r}',\omega)\delta n_v(\mathbf{r}',\omega) + \sum_\alpha f^{q_\alpha}_{Mxc}(\mathbf{r},\omega)\delta q_{\alpha,v}(\omega), \tag{S51}$$

$$\delta v^{(1)}_{\mathrm{KS},j}(\mathbf{r},\omega) = \int d\mathbf{r}' f^{n}_{Mxc}(\mathbf{r},\mathbf{r}',\omega)\delta n_j(\mathbf{r}',\omega) + \sum_\alpha f^{q_\alpha}_{Mxc}(\mathbf{r},\omega)\delta q_{\alpha,j}(\omega), \tag{S52}$$

$$\delta j^{(1)}_{\alpha,\mathrm{KS},v}(\omega) = \int d\mathbf{r} g^{n_\alpha}_{M}(\mathbf{r})\delta n_v(\mathbf{r},\omega), \tag{S53}$$

$$\delta j^{(1)}_{\alpha,\mathrm{KS},j}(\omega) = \delta j_\alpha(\omega) + \int d\mathbf{r} g^{n_\alpha}_{M}(\mathbf{r})\delta n_j(\mathbf{r},\omega). \tag{S54}$$

The mean-field kernel is given by $g^{n_\alpha}_M(\mathbf{r}) = -\omega_\alpha^2 \boldsymbol{\lambda}_\alpha \cdot \mathbf{r}$. As stated above, the subscripts (v, j) on the responses, KS potentials and currents signifies as to what external perturbations $(\delta v(\mathbf{r},t), \delta j_\alpha(t))$ is being considered. The Kohn-Sham scheme of QEDFT decouples the interacting system such that the responses are paired as $(\delta n_v(\mathbf{r},\omega), \delta q_{\alpha,v}(\omega))$ due to $\delta v(\mathbf{r},\omega)$ and $(\delta n_j(\mathbf{r},\omega), \delta q_{\alpha,j_\alpha}(\omega))$ due to $\delta j_\alpha(\omega)$. Therefore, substituting Eqs.(S51) and (S53) into Eqs.(S43)-(S44) and Eqs.(S47)-(S48) and after some simplification, we obtain

$$\sum_{j,b}\left[\delta_{ab}\delta_{ij}\left(\omega_{ai}-\omega\right) + K_{ai,jb}(\omega)\right]\mathbf{P}^{(1)}_{bj,v}(\omega) + K_{ai,bj}(\omega)\mathbf{P}^{(1)}_{jb,v}(\omega) + \sum_\alpha \delta_{ab}\delta_{ij} M_{\alpha,bj}(\omega)\left(\mathbf{L}^{(1)}_{\alpha,v,-}(\omega) + \mathbf{L}^{(1)}_{\alpha,v,+}(\omega)\right)$$
$$= -v_{ai}(\omega), \tag{S55}$$

$$\sum_{j,b}\left[\delta_{ab}\delta_{ij}\left(\omega_{ai}+\omega\right) + K_{ia,bj}(\omega)\right]\mathbf{P}^{(1)}_{jb,v}(\omega) + K_{ia,jb}(\omega)\mathbf{P}^{(1)}_{bj,v}(\omega) + \sum_\alpha \delta_{ab}\delta_{ij} M_{\alpha,jb}(\omega)\left(\mathbf{L}^{(1)}_{\alpha,v,-}(\omega) + \mathbf{L}^{(1)}_{\alpha,v,+}(\omega)\right)$$
$$= -v_{ia}(\omega), \tag{S56}$$

$$\left[\omega_\alpha - \omega\right]\mathbf{L}^{(1)}_{\alpha,v,-}(\omega) + \sum_{jb}\left[N_{\alpha,jb}\mathbf{P}^{(1)}_{bj,v}(\omega) + N_{\alpha,bj}\mathbf{P}^{(1)}_{jb,v}(\omega)\right] = 0, \tag{S57}$$

$$\left[\omega_\alpha + \omega\right]\mathbf{L}^{(1)}_{\alpha,v,+}(\omega) + \sum_{jb}\left[N_{\alpha,jb}\mathbf{P}^{(1)}_{bj,v}(\omega) + N_{\alpha,bj}\mathbf{P}^{(1)}_{jb,v}(\omega)\right] = 0, \tag{S58}$$

Also, substituting Eqs.(S52) and (S54) into Eqs.(S45)-(S46) and Eqs.(S49)-(S50) and after some simplification, we obtain

$$\sum_{j,b}\delta_{ab}\delta_{ij}\left[\left((\omega_{ai}-\omega)+K_{ai,jb}(\omega)\right)\mathbf{P}^{(1)}_{bj,j}(\omega)+K_{ai,bj}(\omega)\mathbf{P}^{(1)}_{jb,j}(\omega)+\sum_{\alpha}M_{\alpha,bj}(\omega)\left[\mathbf{L}^{(1)}_{\alpha,j,-}(\omega)+\mathbf{L}^{(1)}_{\alpha,j,+}(\omega)\right]\right]=0, \tag{S59}$$

$$\sum_{j,b}\delta_{ab}\delta_{ij}\left[((\omega_{ai}+\omega)+K_{ia,bj}(\omega))\mathbf{P}^{(1)}_{jb,j}(\omega)+K_{ia,jb}(\omega)\mathbf{P}^{(1)}_{bj,j}(\omega)+\sum_{\alpha}M_{\alpha,jb}(\omega)\left[\mathbf{L}^{(1)}_{\alpha,j,-}(\omega)+\mathbf{L}^{(1)}_{\alpha,j,+}(\omega)\right]\right]=0, \tag{S60}$$

$$[\omega_\alpha-\omega]\,\mathbf{L}^{(1)}_{\alpha,j,-}(\omega)+\sum_{jb}\left[N_{\alpha,jb}\mathbf{P}^{(1)}_{bj,j}(\omega)+N_{\alpha,bj}\mathbf{P}^{(1)}_{jb,j}(\omega)\right]=-\frac{1}{2\omega_\alpha^2}\delta j_\alpha(\omega), \tag{S61}$$

$$[\omega+\omega_\alpha]\,\mathbf{L}^{(1)}_{\alpha,j,+}(\omega)+\sum_{jb}\left[N_{\alpha,jb}\mathbf{P}^{(1)}_{bj,j}(\omega)+N_{\alpha,bj}\mathbf{P}^{(1)}_{jb,j}(\omega)\right]=-\frac{1}{2\omega_\alpha^2}\delta j_\alpha(\omega), \tag{S62}$$

where we defined the coupling matrices

$$K_{ai,jb}(\omega)=\iint d\mathbf{r}d\mathbf{y}\varphi_i(\mathbf{r})\varphi_a^*(\mathbf{r})f^{n}_{Mxc}(\mathbf{r},\mathbf{y},\omega)\varphi_b(\mathbf{y})\varphi_j^*(\mathbf{y}), \tag{S63}$$

$$M_{\alpha,ai}(\omega)=\int d\mathbf{r}\varphi_i(\mathbf{r})\varphi_a^*(\mathbf{r})f^{q_\alpha}_{Mxc}(\mathbf{r},\omega), \tag{S64}$$

$$N_{\alpha,ia}=\frac{1}{2\omega_\alpha^2}\int d\mathbf{r}\varphi_i^*(\mathbf{r})\varphi_a(\mathbf{r})g^{n_\alpha}_{M}(\mathbf{r}), \tag{S65}$$

and

$$v_{ia}(\omega)=\int d\mathbf{r}\varphi_i^*(\mathbf{r})\delta v(\mathbf{r},\omega)\varphi_a(\mathbf{r}). \tag{S66}$$

The coupling matrix $N_{\alpha,ia}$ has no frequency dependence since this is just the mean-field kernel of the photon modes. We now introduce the following abbreviations $L(\omega)=\delta_{ab}\delta_{ij}\left(\epsilon_a-\epsilon_i\right)+K_{ai,jb}(\omega)$, $K(\omega)=K_{ai,jb}(\omega)$, $M(\omega)=M_{\alpha,bj}(\omega)$, $N=N_{\alpha,bj}$, $\mathbf{X}_1(\omega)=\mathbf{P}^{(1)}_{bj,v}(\omega)$, $\mathbf{Y}_1(\omega)=\mathbf{P}^{(1)}_{jb,v}(\omega)$, $\mathbf{X}_2(\omega)=\mathbf{P}^{(1)}_{bj,j}(\omega)$, $\mathbf{Y}_2(\omega)=\mathbf{P}^{(1)}_{jb,j}(\omega)$, $\mathbf{A}_1(\omega)=\mathbf{L}^{(1)}_{\alpha,v,-}(\omega)$, $\mathbf{B}_1(\omega)=\mathbf{L}^{(1)}_{\alpha,v,+}(\omega)$, $\mathbf{A}_2(\omega)=\mathbf{L}^{(1)}_{\alpha,j,-}(\omega)$, $\mathbf{B}_2(\omega)=\mathbf{L}^{(1)}_{\alpha,j,+}(\omega)$, $V(\omega)=-v_{ai}(\omega)$, $J_\alpha(\omega)=-\frac{\delta j_\alpha(\omega)}{2\omega_\alpha^2}$.

Using these notations, we cast Eqs.(S55)-(S58) and Eqs.(S59)-(S62) into two matrix equations given by

$$\left[\begin{pmatrix} L(\omega) & K(\omega) & M(\omega) & M(\omega) \\ K^*(\omega) & L(\omega) & M^*(\omega) & M^*(\omega) \\ N & N^* & \omega_\alpha & 0 \\ N & N^* & 0 & \omega_\alpha \end{pmatrix} + \omega \begin{pmatrix} -1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}\right] \begin{pmatrix} \mathbf{X}_1(\omega) \\ \mathbf{Y}_1(\omega) \\ \mathbf{A}_1(\omega) \\ \mathbf{B}_1(\omega) \end{pmatrix} = \begin{pmatrix} V(\omega) \\ V^*(\omega) \\ 0 \\ 0 \end{pmatrix} \tag{S67}$$

$$\left[\begin{pmatrix} L(\omega) & K(\omega) & M(\omega) & M(\omega) \\ K^*(\omega) & L(\omega) & M^*(\omega) & M^*(\omega) \\ N & N^* & \omega_\alpha & 0 \\ N & N^* & 0 & \omega_\alpha \end{pmatrix} + \omega \begin{pmatrix} -1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}\right] \begin{pmatrix} \mathbf{X}_2(\omega) \\ \mathbf{Y}_2(\omega) \\ \mathbf{A}_2(\omega) \\ \mathbf{B}_2(\omega) \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ J_\alpha(\omega) \\ J_\alpha(\omega) \end{pmatrix} \tag{S68}$$

Next, we argue that the right hand side of the above matrices remains finite as the frequency ω approaches the exact excitation frequencies $\omega \to \Omega_q$ of the interacting system while the density and displacement field responses on the left hand side has poles at the true excitation frequencies Ω_q. This allows us to cast Eq. (S67) and Eq. (S68) into an eigenvalue problem

$$\begin{pmatrix} L(\Omega_q) & K(\Omega_q) & M(\Omega_q) & M(\Omega_q) \\ K^*(\Omega_q) & L(\Omega_q) & M^*(\Omega_q) & M^*(\Omega_q) \\ N & N^* & \omega_\alpha & 0 \\ N & N^* & 0 & \omega_\alpha \end{pmatrix} \begin{pmatrix} \mathbf{X}_1(\Omega_q) \\ \mathbf{Y}_1(\Omega_q) \\ \mathbf{A}_1(\Omega_q) \\ \mathbf{B}_1(\Omega_q) \end{pmatrix} = \Omega_q \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix} \begin{pmatrix} \mathbf{X}_1(\Omega_q) \\ \mathbf{Y}_1(\Omega_q) \\ \mathbf{A}_1(\Omega_q) \\ \mathbf{B}_1(\Omega_q) \end{pmatrix} \tag{S69}$$

$$\begin{pmatrix} (\Omega_q) & K(\Omega_q) & M(\Omega_q) & M(\Omega_q) \\ K^*(\Omega_q) & L(\Omega_q) & M^*(\Omega_q) & M^*(\Omega_q) \\ N & N^* & \omega_\alpha & 0 \\ N & N^* & 0 & \omega_\alpha \end{pmatrix} \begin{pmatrix} \mathbf{X}_2(\Omega_q) \\ \mathbf{Y}_2(\Omega_q) \\ \mathbf{A}_2(\Omega_q) \\ \mathbf{B}_2(\Omega_q) \end{pmatrix} = \Omega_q \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix} \begin{pmatrix} \mathbf{X}_2(\Omega_q) \\ \mathbf{Y}_2(\Omega_q) \\ \mathbf{A}_2(\Omega_q) \\ \mathbf{B}_2(\Omega_q) \end{pmatrix} \tag{S70}$$

It is convenient to cast Eqs.(S69) and (S70) into a Hermitian eigenvalue problem which is given by

$$\begin{pmatrix} U & V \\ V^T & \omega_\alpha^2 \end{pmatrix} \begin{pmatrix} \mathbf{E}_1 \\ \mathbf{P}_1 \end{pmatrix} = \Omega_q^2 \begin{pmatrix} \mathbf{E}_1 \\ \mathbf{P}_1 \end{pmatrix}, \tag{S71}$$

$$\begin{pmatrix} U & V \\ V^T & \omega_\alpha^2 \end{pmatrix} \begin{pmatrix} \mathbf{E}_2 \\ \mathbf{P}_2 \end{pmatrix} = \Omega_q^2 \begin{pmatrix} \mathbf{E}_2 \\ \mathbf{P}_2 \end{pmatrix}, \tag{S72}$$

where we assumed real-valued orbitals, i.e., $K = K^*$, $M = M^*$ and $N = N^*$, and the matrices are given by $U = (L-K)^{1/2}(L+K)(L-K)^{1/2}$, $V = 2(L-K)^{1/2}M^{1/2}N^{1/2}\omega_\alpha^{1/2}$, $V^* = 2\omega_\alpha^{1/2}N^{1/2}M^{1/2}(L-K)^{1/2}$, and the eigenvectors are $\mathbf{E}_1 = N^{1/2}(L-K)^{-1/2}(\mathbf{X}_1 + \mathbf{Y}_1)$ and $\mathbf{P}_1 = M^{1/2}\omega_\alpha^{-1/2}(\mathbf{A}_1 + \mathbf{B}_1)$.

The pseudo-eigenvalue problem of Eqs.(S71) and (S72) is the final form of QEDFT matrix equation for obtaining exact excitation frequencies and oscillator strengths.

S5 Oscillator Strengths

In this section, we derive the oscillator strengths resulting from the eigenvectors of the pseudo-eigenvalue problem of Eqs.(S71) and (S72). Multiplying out Eq. (S67), we write the matrix equation in the form

$$\begin{aligned}
(L+K)(\mathbf{X}_1+\mathbf{Y}_1)+2M(\mathbf{A}_1+\mathbf{B}_1)-\omega(\mathbf{X}_1-\mathbf{Y}_1) &= -2\boldsymbol{v},\\
(L-K)(\mathbf{X}_1-\mathbf{Y}_1)-\omega(\mathbf{X}_1+\mathbf{Y}_1) &= 0,\\
2N(\mathbf{X}_1+\mathbf{Y}_1)+\omega_\alpha(\mathbf{A}_1+\mathbf{B}_1)-\omega(\mathbf{A}_1-\mathbf{B}_1) &= 0,\\
\omega_\alpha(\mathbf{A}_1-\mathbf{B}_1)-\omega(\mathbf{A}_1+\mathbf{B}_1) &= 0.
\end{aligned}$$

From here on we set $S=(L-K)$, the above pair of equations now becomes

$$\begin{aligned}
&S(L+K)\mathbf{E}_1+2SM\mathbf{P}_1-\omega^2\mathbf{E}_1=-2S\boldsymbol{v},\\
&2\omega_\alpha N\mathbf{E}_1+\omega_\alpha^2\mathbf{P}_1-\omega^2\mathbf{P}_1=0.
\end{aligned}$$

This can be written in matrix form as

$$\left[\begin{pmatrix} S(L+K) & 2SM \\ 2\omega_\alpha N & \omega_\alpha^2 \end{pmatrix} - \omega^2\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}\right]\begin{pmatrix} \mathbf{E}_1 \\ \mathbf{P}_1 \end{pmatrix} = -\begin{pmatrix} 2S\boldsymbol{v} \\ 0 \end{pmatrix}, \tag{S73}$$

where $\mathbf{E}_1=\mathbf{X}_1+\mathbf{Y}_1$ and $\mathbf{P}_1=\mathbf{A}_1+\mathbf{B}_1$. We perform the same steps as above to make the nonlinear eigenvalue problem Hermitian and obtain

$$\left[C-\omega^2\mathbb{1}\right]\begin{pmatrix} N^{1/2}S^{-1/2}\mathbf{E}_1 \\ M^{1/2}\omega_\alpha^{-1/2}\mathbf{P}_1 \end{pmatrix} = -\begin{pmatrix} 2N^{1/2}S^{1/2}\boldsymbol{v} \\ 0 \end{pmatrix}, \tag{S74}$$

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

$$\mathbf{E}_1=-2S^{1/2}\left[C-\omega^2\mathbb{1}\right]^{-1}S^{1/2}\boldsymbol{v}, \tag{S75}$$

$$\mathbf{P}_1=-2\omega_\alpha^{1/2}M^{-1/2}\left[C-\omega^2\mathbb{1}\right]^{-1}N^{1/2}S^{1/2}\boldsymbol{v}. \tag{S76}$$

When $\mathbf{Z}_I$ is normalized, we can use the spectral expansion to get

$$\left[C - \omega^2 \mathbb{1}\right]^{-1} = \sum_I \frac{\mathbf{Z}_I \mathbf{Z}_I^\dagger}{\Omega_I^2 - \omega^2}, \tag{S77}$$

where $\mathbf{Z}_I = \begin{pmatrix} \mathbf{E}_{1I} \\ \mathbf{P}_{1I} \end{pmatrix}$. The oscillator strength for the density-density response function which is related to the dynamic polarizability is given in Eq.(54) in the main manuscript.

S5.1 Oscillator strength for the photon-matter response function

Next, we substitute the expression of the spectral expansion Eq. (S77) in Eq. (S76) and by substituting $\mathbf{P}_1$ in Eq. (S40) yields

$$\delta q_{\alpha,v}(\omega) = \sum_I \left\{ \frac{2\omega_\alpha^{1/2} M^{-1/2} \mathbf{Z}_I \mathbf{Z}_I^\dagger N^{1/2} S^{1/2}}{\omega^2 - \Omega_I^2} \right\} v(\omega).$$

The oscillator strength is given by

$$f_{I,\alpha}^{pn} = 2\omega_\alpha^{1/2} M^{-1/2} \mathbf{Z}_I \mathbf{Z}_I^\dagger N^{1/2} S^{1/2}. \tag{S78}$$

Also, from Eq.(36) of the main manuscript and using the Lehmann representation of the response function $\chi_n^{q_\alpha}(\mathbf{r}', \omega)$ the response $\delta q_{\alpha,v}(\omega)$ is given by

$$\delta q_{\alpha,v}(\omega) = \int d\mathbf{r}' \sum_k \left[\frac{2\Omega_k \langle \Psi_0 | \hat{q}_\alpha | \Psi_k \rangle \langle \Psi_k | \hat{n}(\mathbf{r}') | \Psi_0 \rangle}{\omega^2 - \Omega_k^2} \right] \delta v(\mathbf{r}', \omega),$$

The oscillator strength of Eq.(S78) can be expressed as matrix elements of the internal pair $(\hat{n}(\mathbf{r}), \hat{q}_\alpha)$ as

$$f_{\alpha,k}(\mathbf{r}') = 2\Omega_k \langle \Psi_0 | \hat{q}_\alpha | \Psi_k \rangle \langle \Psi_k | \hat{n}(\mathbf{r}') | \Psi_0 \rangle \equiv f_{I,\alpha}^{pn}. \tag{S79}$$

S5.2 Oscillator strength for the matter-photon response function

Following similar steps as above with Eq. (S68) we obtain

$$\mathbf{E}_2 = -2S^{1/2}N^{-1/2}\left[C-\omega^2\mathbb{1}\right]^{-1}M^{1/2}\omega_\alpha^{1/2}J'_\alpha, \tag{S80}$$

$$\mathbf{P}_2 = -2\omega_\alpha^{1/2}\left[C-\omega^2\mathbb{1}\right]^{-1}\omega_\alpha^{1/2}J'_\alpha. \tag{S81}$$

where $J'_\alpha(\omega) = \frac{j_\alpha(\omega)}{2\omega_\alpha^2}$ and $J_\alpha(\omega) = -J'_\alpha(\omega)$. By substituting the spectral expansion Eq. (S77) in $\mathbf{E}_2$ and further substituting in Eq. (S41) yields

$$\delta n_j(\mathbf{r},\omega) = -2\sum_{ia,I}\frac{\Phi_{ia}S^{1/2}N^{-1/2}\mathbf{Z}_I\mathbf{Z}_I^\dagger M^{1/2}\omega_\alpha^{1/2}\Phi_{ai}}{(\Omega_I^2-\omega^2)}J'_\alpha(\omega).$$

Following a similar procedure as above, we express the density response to the external charge current as

$$\delta n_j(\mathbf{r},\omega) = \sum_I\left\{\frac{\Phi_{ia}S^{1/2}N^{-1/2}\mathbf{Z}_I\mathbf{Z}_I^\dagger M^{1/2}\omega_\alpha^{1/2}\Phi_{ia}}{\omega^2-\Omega_I^2}\right\}\frac{j_\alpha(\omega)}{\omega_\alpha^2},$$

where $\Phi_{ia}(\mathbf{r}) = \varphi_i^*(\mathbf{r})\varphi_a(\mathbf{r})$ and the oscillator strength is given by

$$f_{I,\alpha}^{np} = \frac{1}{\omega_\alpha}\Phi_{ia}S^{1/2}N^{-1/2}\mathbf{Z}_I\mathbf{Z}_I^\dagger M^{1/2}\omega_\alpha^{1/2}\Phi_{ia}. \tag{S82}$$

From Eq.(37) of the main manuscript and using the Lehmann representation of the response function $\chi_{q_\alpha}^n(\mathbf{r},\omega)$, the response $\delta n_j(\mathbf{r},\omega)$ is given by

$$\delta n_j(\mathbf{r},\omega) = \sum_{\alpha,k}\left[\frac{2\Omega_k\langle\Psi_0|\hat{n}(\mathbf{r})|\Psi_k\rangle\langle\Psi_k|\hat{q}_\alpha|\Psi_0\rangle}{\omega^2-\Omega_k^2}\right]\frac{\delta j_\alpha(\omega)}{\omega_\alpha},$$

The oscillator strength of Eq.(S82) can be expressed as matrix elements of the internal pair $(\hat{n}(\mathbf{r}),\hat{q}_\alpha)$ as

$$f_{k,\alpha}(\mathbf{r}) = 2\Omega_k\langle\Psi_0|\hat{n}(\mathbf{r})|\Psi_k\rangle\langle\Psi_k|\hat{q}_\alpha|\Psi_0\rangle \equiv f_{I,\alpha}^{np}. \tag{S83}$$

S5.3 Oscillator strength for the photon-photon response function

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

$$\delta Q(t) = \sum_\alpha \delta q_\alpha(t). \tag{S84}$$

The field polarizability in frequency space can be written as

$$\beta_\alpha(\omega) = \sum_{\alpha'} \frac{\delta q_\alpha(\omega)}{\delta j_{\alpha'}(\omega)}. \tag{S85}$$

By substituting Eq. (S81) in Eq. (S42) and using the spectral expansion yields

$$\delta q_{\alpha,j}(\omega) = -\sum_I \frac{2\omega_\alpha^{1/2}\mathbf{Z}_I\mathbf{Z}_I^\dagger\omega_\alpha^{1/2}}{\Omega_I^2 - \omega^2} J'_\alpha.$$

By substituting the above relation in Eq. (S85) we obtain

$$\beta_\alpha(\omega) = -\sum_{\alpha'}\sum_I \frac{2\omega_\alpha^{1/2}\mathbf{Z}_I\mathbf{Z}_I^\dagger\omega_\alpha^{1/2}}{\Omega_I^2 - \omega^2} \frac{\delta j_\alpha(\omega)/2\omega_\alpha^2}{\delta j_{\alpha'}(\omega)},$$

which simplifies to

$$\beta_\alpha(\omega) = -\sum_I \frac{1}{\omega_\alpha^2} \frac{\omega_\alpha^{1/2}\mathbf{Z}_I\mathbf{Z}_I^\dagger\omega_\alpha^{1/2}}{\Omega_I^2 - \omega^2}. \tag{S86}$$

Eq. (S86) is the field polarizability analogous to the atomic polarizability tensor of Eq. (52) of the main manuscript. As in Eq.(53) of the main manuscript in which the molecular isotropic polarizability, $\alpha(\omega)$ is defined as the mean value of three diagonal elements of the polarizability tensor, i.e., $\alpha(\omega) = 1/3\left(\alpha_{xx}(\omega) + \alpha_{yy}(\omega) + \alpha_{zz}(\omega)\right)$, we analogously define an absorption cross section of the field given by

$$\tilde{\sigma}_{\alpha}(\omega) \equiv \frac{4\pi\omega}{c}\mathcal{I}m\ \mathrm{Tr}\beta_{\alpha}(\omega)/3. \tag{S87}$$

For the oscillator strength, from Eq.(38) of the main manuscript and using the Lehmann representation of the response function $\chi^{q_{\alpha}}_{q_{\alpha'}}(\omega)$ the response $\delta q_{\alpha,j}(\omega)$ is given by

$$\delta q_{\alpha,j}(\omega) = \sum_{\alpha',k}\left[\frac{2\Omega_k\langle\Psi_0|\hat{q}_{\alpha}|\Psi_k\rangle\langle\Psi_k|\hat{q}_{\alpha'}|\Psi_0\rangle}{\omega^2 - \Omega_k^2}\right]\frac{\delta j_{\alpha'}(\omega)}{\omega_{\alpha'}}.$$

We find the oscillator strength

$$f^{pp}_{I,\alpha} = \frac{1}{3\omega_{\alpha}^2}\left|\mathbf{Z}_I^{\dagger}\omega_{\alpha}^{1/2}\right|^2 = \frac{2}{3}\Omega_I\sum_{\alpha'}\frac{1}{\omega_{\alpha'}}\langle\Psi_0|\hat{q}_{\alpha}|\Psi_I\rangle\langle\Psi_I|\hat{q}_{\alpha'}|\Psi_0\rangle. \tag{S88}$$

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