Time-dependent density-functional theory (TDDFT) assumes systems as perfectly isolated, providing discrete resonances with infinite lifetime. We derive a simple local potential that embeds TDDFT into a photonic continuum, taking the form of radiation-reaction forces. This self-consistently provides access to radiative emission, natural linewidth, mass renormalization, electromagnetically induced transparency, Purcell-enhanced and superradiant emission. The seamless integration into existing TDDFT libraries with virtually no additional cost paves the way for new perspectives in TDDFT and *ab initio* QED.

**RADIATION-REACTION POTENTIAL DESCRIBING RADIATIVE EMISSION**

Let us assume for a moment that the quantum-nature of light is not essential for our endeavor – we will indeed recover many from QED expected predictions. The non-relativistic dynamic of condensed matter is then governed by the minimally coupled Coulomb Hamiltonian

\[ \hat{H} = \sum_i \frac{1}{2m_i} (-i\hbar \nabla_i - q_i \mathbf{A}(\mathbf{r}_i,t)/c)^2 + \hat{H}_\parallel + \varepsilon_0/2 \int d\mathbf{r} |\mathbf{E}_\perp(\mathbf{r})|^2 + c^2 |\mathbf{B}(\mathbf{r})|^2 \]

with fixed Coulomb gauge \( \nabla \cdot \mathbf{A} = 0 \). The electromagnetic fields obey Maxwell’s equation of motion and couple self-consistently with the Schrödinger equation which describes our matter system of interest. The internal structure of the latter, consisting out of electrons and nuclei, is determined by the longitudinal Coulombic interactions \( \hat{H}_\parallel = 1/(8\pi\varepsilon_0) \sum_{i,j} N_e N_n q_i q_j/|\mathbf{r}_i - \mathbf{r}_j| \), here given in free-space.

The transverse electric \( \mathbf{E}_\perp(\mathbf{r}) = -1/c\partial_t \mathbf{A}(\mathbf{r}) \) and magnetic fields \( \mathbf{B}(\mathbf{r}) = 1/c\nabla \times \mathbf{A}(\mathbf{r}) \) are in contrast not bound to the material but can propagate into free-space. If the wavelength of those propagating fields is substantially larger than the matter system, the spatial dependence of the transverse fields is commonly neglected. Even in this strongly simplified limit, the remaining task of solving the Schrödinger equation is extraordinary challenging.

Time-dependent density-functional theory evolves around the concept that effective single-particle Kohn-Sham equations exist that are able to uniquely mimic all physical observables which are inherited by the original Coulomb Hamiltonian [33, 34, 48]. Our goal is now to provide a local potential that is as simple as possible to describe light-matter coupling and radiative emission.
consistent with the above Hamiltonian within the ordinary
time-dependent Kohn-Sham equations

\[ i\hbar \partial_t \phi_i(\mathbf{r}t) = \left( -\frac{\hbar^2}{2m_e} \nabla^2 + v_s(\mathbf{r}t) \right) \phi_i(\mathbf{r}t) \]

where the local Kohn-Sham potential \( v_s = v_{\text{ext}} + v_{\text{Hxc}} + v_{\text{rr}} \) consists of the external potential \( v_{\text{ext}} \) (usually the nuclear binding potential), the electronic Hartree exchange-correlation potential \( v_{\text{Hxc}} \) mimicking electronic many-body interactions, and the potential \( v_{\text{rr}} \) that will account for the emission of light.

Assuming external fields will merely drive the matter-system, it is standard in TDDFT to apply the local electronic potential \( v_{\text{drive}}(\mathbf{r}t) = e\mathbf{r} \cdot \mathbf{E}_{\text{drive}}(t) \). Naturally, driving the system externally will lead to absorption and excitation. Let us turn this idea now around in order to describe the emission of light on equal footing. Each current will induce an electromagnetic field for which its precise spatial and polarization structure depends on the electromagnetic environment – oscillating charges emit light. The generated field can be expressed with the help of the dyadic Green’s tensor

\[ \mathbf{E}_r(\mathbf{r}, \omega) = i\mu_0\omega \int_V d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot (-e\mathbf{j}(\mathbf{r}', \omega)) \]  

(1)

which is the formal solution of Helmholtz’s equation [49]

\[ \left[ \nabla \times \frac{1}{\mu_r(\omega)} \nabla \times -\omega^2\mu_0\varepsilon_0\varepsilon_r(\omega) \right] \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') \]

including linear media \( \varepsilon_r, \mu_r \), where clearly the choice of gauge has to be consistent with the TDDFT description. The energy associated with the radiated field should be taken correctly from the electronic system, i.e., the system of oscillating charges should feel a recoil force that accounts for any emitted energy – the radiation reaction, ensuring Newton’s third law. Taking inspiration from the above driving potential, we can intuitively conclude that an additional potential, the (dipolar) radiation-reaction potential \( v_{\text{rr}}(\mathbf{r}t) = e\mathbf{r} \cdot \mathbf{E}_r(\mathbf{r}, \omega) \), should be added to the ordinary Kohn-Sham equations which accounts for the loss of energy due to photonic emission \( \Delta E_{\text{rr}}(t) = \int_{t_0}^t dt' \mathbf{E}_r(t') \cdot \mathbf{E}_{r,\perp}(t) \). Where, fixing the nuclei for the moment, \( \mathbf{R}(t) = \int dt'(e\mathbf{r}(t')) \) is the expectation value of the electronic dipole moment. The recoil force augments the light-matter coupling with self-consistency and manifests itself for instance as ‘jerk’ \( \ddot{x}(t) \) in the classical Abraham-Lorentz model [50], elaborated in more detail in the SI. For illustration, let us assume a single freespace dimension (\( \varepsilon_r = \mu_r = 1 \)) for the photonic fields with periodic boundaries, i.e., the matter-system emits into a waveguide. The eigenmodes of Helmholtz’s equation are then \( S(\mathbf{r}) = \sqrt{1/\nabla^2[\cos(kx) + \sin(kx)])} \), \( k = \frac{2\pi n_x}{L_x} \), \( n_x \in \mathbb{Z} \) and we enforce the transverse polarization \( \varepsilon_x \perp \mathbf{k} \). The Helmholtz equation in Coulomb gauge can be solved by usage of the spectral theorem

\[ \mathbf{G}(x, x', \omega) = \sum_{\mathbf{k}} \frac{S(\mathbf{k})S(\mathbf{r}/\omega)T}{k^2 - (\omega/c)^2} \mathbf{e}_x^T. \]

In combination with the inverse Fourier transformation and performing the consistent long-wavelength approximation \( (x = x' = 0) \), details in the SI), the radiation-reaction potential takes the form \( v_{\text{rr}}(\mathbf{r}t) = e\mathbf{r} \cdot \varepsilon_x \sum_{\mathbf{k}} \frac{1}{v_{ce}} \int_0^t dt' \cos(ck(t - t')) \int_V d\mathbf{r}' \mathbf{e}_x \cdot (-e\mathbf{j}(\mathbf{r}', t')) \) and we notice that \( v_{\text{rr}} \propto e_x \cdot \mathbf{r} e_e \cdot (\mathbf{r}' t'), \) i.e., the transverse projection of the current induces a (memory dependent) recoil acting back on the electronic system, as we envisioned. [51]

When the number of photonic modes is increased, the wide and dense mode-structure will start to represent a photonic bath. Performing the explicit integration and employing the continuity equation for the integrated current \( \int \mathbf{e}_x(t') \) we obtain the TDDFT radiation-reaction potential accounting for the recoil forces of emitting into a waveguide

\[ v_{\text{rr}}^{1D}(\mathbf{r}t) = \frac{4\pi \hbar c}{e^2} A^{-1} \mathbf{e}_c \cdot \mathbf{R}(t) \mathbf{e}_c \cdot (-e\mathbf{r}), \]  

(2)

i.e., the moving charges will be slowed down by an always repelling potential. The coupling between the photonic continuum (representing the perfect wide-band limit) and the electronic system is provided by the fine-structure constant \( \alpha \), as intuitively expected, divided by the cross-sectional area of the waveguide \( A = V/L_x \). The integrated emitted energy takes the form \( \Delta E_{\text{rr}}(t) = \frac{4\pi \hbar c}{e^2} A^{-1} \int_{t_0}^t dt' \mathbf{e}_c \cdot \mathbf{R}(t')^2 \). While we derived \( v_{\text{rr}} \) based on radiation-reaction forces, we can alternatively perform the explicit limit from quantum electrodynamics to obtain the same result (see the SI for a detailed derivation).

While ångström thickness waveguides are possible [52], the majority of interesting systems (such as typical Fabry-Perot-type cavities) will feature comparably large quantization volumes, leading to a small influence of the radiation-reaction on the dynamics. Plasmonic systems represent here an exception due to their large currents and high mode-density at nanometer scales [53–55] which substantially enhances the emission of nearby molecular systems (Purcell-enhancement) as discussed later. An extended discussion and first generalizations of the radiation-reaction potential to emission near mirrors and full 3D can be found in the SI, we will remain here with the form introduced in eq. (2).

Let us illustrate in the following the broad impact of the radiation-reaction potential despite its simplicity. Figure 1 presents the quick radiative decay of hydrogen driven by an external pulse.

External perturbations \( \delta v_{\text{appl}} \) induce a response of the system which results in electronic motion and thus emission according to the radiation-reaction forces. In linear order, the response of the electronic density is given by

\[ \delta \rho(\mathbf{r}t) = \int \mathbf{d}r'dt' \chi_{s}(\mathbf{r}, \mathbf{r}', t - t') \delta v_{\text{appl}}(\mathbf{r}' t') + \int \mathbf{d}r' dt'' f(\mathbf{r}, \mathbf{r}', t' - t'') \delta \rho(\mathbf{r}' t''). \]

Here \( f(\mathbf{r}, \mathbf{r}', t' - t'') \) is defined in the SI for the linear response of the Kohn-Sham potential (which depends self-consistently on the density) when the electronic
Figure 1. Dipole $R(t)$ (black solid), relative electronic energy $\Delta E_e$ (blue dashed) and accumulated emitted radiation-reaction energy $\Delta E_{rr}$ (red dashed-dotted) for one-dimensional soft-coulomb hydrogen $v_{ext}(x) = e^2/(4\pi \varepsilon_0 \sqrt{x^2 + 1})$ emitting into a waveguide assuming a strongly Purcell-enhanced emission with $A^{-1} = 1/\alpha_0^2$ for illustrative reasons. Notice that Purcell-enhancement factors of order $\Gamma/\Gamma_0 \approx O(10^5)$ are not unusual. (a) An external laser-pulse $E_{ext}(t)$ (yellow, solid, amplified by a factor 10) drives the system out of its ground-state. Energy deposited in the electronic system by the pulse is quickly radiated into the photonic bath. (b) Stimulated emission from the first excited electronic state under external driving. Details in SI.

Density is perturbed. The contribution of the radiation-reaction forces $f_{RR}^{1D}(r', r'', \omega) = i4\pi\hbar A^{-1}\omega e_c \cdot r' e_c \cdot r''$ is explicitly memory- and frequency-dependent which allows it to provide a broadened resonance in contrast to the widely used frequency-independent adiabatic kernels that demand ad hoc broadening by hand. If we assume a single occupied $(g)$ and a single unoccupied $(e)$ Kohn-Sham orbital with bare excitation energy $\Omega_{eg} = \hbar \omega_{eg} = \varepsilon_e - \varepsilon_g$, we can solve the linear-response Casida equation analytically to obtain the excitation poles $\Omega_n = \pm \Omega_{eg} \sqrt{1 - (4\pi \alpha A^{-1}/e^2)^2|e_c \cdot R_{eg}|^2 + i(4\pi \alpha A^{-1}/e^2)|e_c \cdot R_{eg}|^2}$ and the polarizability tensor [56] defined by $R_{induced}(\omega) = \alpha(\omega) \cdot E_{perturbation}(\omega)$ as

$$\alpha_{\mu\nu}(\omega) = \sum_{n=1}^{\infty} 2R_{rg}^{(\mu)} R_{ng}^{(\nu)} (\hbar \omega - \Omega_n)^2 + (3\Omega_n)^2 = 2R_{ge}^{(\mu)} R_{eg}^{(\nu)} (\hbar \omega - \Omega_n^2)^2 + \hbar^2 \Gamma_{rr}^2$$

with photoabsorption cross-section and linewidth

$$\sigma(\omega) = \frac{4\pi \omega}{c} \Im(\alpha(\omega)) \quad \Gamma_{rr} = \frac{4\pi \omega \omega_{eg} A^{-1}|e_c \cdot R_{eg}|^2}{e^2}.$$

A system has therefore no longer discrete excitations as common in TDDFT but features a physical linewidth $\Gamma_{rr}$ which is for this simple electromagnetic environment indeed identical to the Wigner-Weisskopf solution $\Gamma_{W,W}^D = \omega_{eg}|e_c \cdot R_{eg}|^2 A^{-1}/\hbar \varepsilon_0 c = \Gamma_{rr}$. This analogy holds only in the perturbative limit, i.e., when the emission process influences the lifetimes they naturally deviate from the perturbative Wigner-Weisskopf prediction (see SI).

The radiation-reaction leads to a slight shift of the resonance as a consequence of the electromagnetic recall $\propto 1 - (4\pi \alpha A^{-1}/e^2|e_c \cdot R_{eg}|^2)^2/2$ (assuming the recoil to be comparably small). This Lamb-shift-like effect indicates that the mass of the particle is changed by the recoil which results in the physical mass of an electron. We therefore found here a tool to obtain access to the Lamb-shift in ordinary TDDFT simulations. For Purcell-enhanced emission in a waveguide, the Lamb-shift can take non-negligible values, especially when collective effects lead to collective Lamb-shift effects [57]. However, for the vast majority of applications this shift is negligible and aspects such as the quality of the exchange-correlation potential in TDDFT are far more influential.

Let us point out that clearly also $ab$ initio QED, QEDFT and plasmonic strong coupling greatly benefit from the radiation-reaction approach. Typically, only a few cavity modes play a substantial role in the strong coupling and the manifold of weakly interacting modes describe the emission profile, i.e., the loss-channels, of the cavity. Using the radiation-reaction potential which represents a bath of free-space modes, cavity losses into free-space can be efficiently modeled as we will see in the following. Such a separation is of imminent importance for the future development of $ab$ initio QED as it allows us to utilize higher level descriptions for a few most relevant cavity modes which explicitly account for the quantum features of light (see e.g. [29]) while retaining the full manifold of modes which account on a simplified level for emission, linewidth and loss. In this sense, the radiation-reaction potential acts as highly efficient realization of a bath, comparable to open-system strategies in quantum optics.

Electromagnetically induced transparency in TDDFT

One excellent example of the strength of such a photonic-bath treatment is the description of electromagnetically induced transparency (EIT) [39, 58]. Let us couple a single loss-free cavity mode, representing for instance a whispering-gallery mode, strongly to the hydrogen system. In addition, the matter system can emit light into a waveguide, i.e., a bath of photonic modes according to eq. (2). Fig. 2 presents how the absorption-profile of hydrogen changes with increasing strength of the bath and the coupling strength to the single mode. The interplay between the high-quality cavity mode and the lossy electromagnetic bath induces a window of transparency for strong radiative emission, i.e., the natural linewidth (black, dashed) obtains a sharp window of transparency (yellow, solid) at which the system can no longer absorb light. EIT has a plethora of technological applications, including effectively stopping or storing light [59, 60], which become now for the first time available to TDDFT and QEDFT in a
simple and intuitive way. EIT emerges from the interplay between low and high quality resonators [58], adjusting free-space emission and quality of the single optical mode allows to transition between EIT and strong coupling.

While it is common practice to describe superradiant effects with quantum optical models [37] and the Purcell enhancement of spontaneous emission as perturbative correction to Wigner-Weisskopf theory [54], we show here that the radiation-reaction potential equips TDDFT with the necessary tools to address those aspects self-consistently. In order to describe realistic systems, we implemented the radiative-reaction potential introduced in eq. (2) in the TDDFT code GPAW [61] using the computationally efficient LCAO basis [62].

### Superradiant linewidth

The synchronized evolution among a set of atoms or molecules amplifies their interaction with the photonic environment. Superradiance describes the effect that the synchronized emission is quicker than the individual emission [37]. For the single-photon absorption spectrum this takes the form of a linearly increasing emission rate with the size of the ensemble and is illustrated in fig. 3 for a set of Sodium dimers. The decisive strength of the ab initio implementation is the consistent treatment of radiative emission and matter, i.e., any form of Coulomb mediated interaction or charge-migration is treated self-consistently. This becomes particularly relevant in situations in which the material has a complicated excitation structure or charge-migration between subsystems appears, e.g., observed in nanoplasmonics [63]. The construction of simplified few-level systems or entirely classical description is then obstructed.

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**Plasmonic Purcell-enhancement**

Plasmonic particles contribute in two ways to the quick decay of nearby molecules. First, strong dipolar oscillations in clusters consistent with the long wavelength approximation (localized surface plasmons) result in quick radiative decay. Second and often dominant, they feature very quick internal dephasing on the fs timescale due to Landau damping and electron-electron scatterings. In combination with charge migration and hot-electron transfer, this results in a complex dynamics which is theoretically challenging to capture. Full-fledged ab initio approaches provide hereby valuable results and following the radiation-reaction, we can now easily and consistently account for radiative features. Fig 4 illustrates the dipolar spectrum of benzene next to Al201 [17] including and excluding the radiative-emission. The strong longitudinal fields around the plasmon lead to a hybridization of the bare plasmonic and \( \pi - \pi^* \) benzene excitation. For small cross-sectional surfaces \( A \), the strong plasmonic currents provide the previously slow radiating benzene excitation with an efficient emission channel – Purcell-enhancement. The larger \( A \) the stronger internal dephasings compete with radiation, up to the point (\( A^{-1} < 10^{-3} \AA^{-2} \)) where the Purcell-enhancement for the emission of benzene is dominated by those internal losses. [64]

The competition between radiative and non-radiative decay channels is often tilted in favor of internal losses while nanoantenna design can invert this characteristic [65, 66]. The generic dipolar form \( v_{rr+es}(\mathbf{r}_t) = e \mathbf{r} \cdot \mathbf{E}_r(\mathbf{r}_t) \), accounting for radiation-reaction and ‘emitted energy’ via electrostatic components, naturally allows for a classical
electromagnetic treatment of (surface) plasmonic excitations. Our ansatz is formally equivalent to the combined propagation of Maxwell and Kohn-Sham system [44], albeit being substantially easier to analytically modify following e.g. Mie-theory and numerically integrate into existing TDDFT libraries. The radiation-reaction potential represents therefore a convenient way to extend existing techniques [67] in which the instantaneous longitudinal plasmonic fields are treated via classical electrodynamics. Such mixed quantum-classical simplifications lose naturally the consistent access to charge-migration, a trade-off that should be evaluated on a case-by-case study. The radiation-reaction ansatz illustrates furthermore how collective strong coupling, which obtained recent interest in QED chemistry [68], can be effectively integrated into TDDFT. In particular, the local response $\delta \rho(r, \omega)$ on a single molecule is perturbed by the effective fields $E_{\text{drive}}(\omega) + E_{\text{r}}(r, \omega)$ which in turn are modified by the large ensemble of collectively radiating molecules, an approach that will be presented in a forthcoming publication.

CONCLUSION

By treating a continuum of transverse electromagnetic modes implicitly, we have been able to derive a simple and elegant local potential for time-dependent density-functional theory accounting for radiative effects. Our local potential represents the TDDFT analogue of radiation-reaction and the Abraham-Lorentz model, i.e., it represents the recoil emerging as a consequence of energy conservation during the emission of light. This radiation-reaction potential provides with virtually no additional effort natural linewidths, radiative damping, electromagnetically induced transparency, Purcell enhancement and superradiant emission from first principles. While we avoid the need to keep track of the photonic fields, the generated fields are determined at any point in time by the dyadic Greens tensor and the electronic currents, allowing a precise description of the experimentally measurable fields. Our conceptions can be extended to the nuclear degrees of freedom, beyond the dipolar approximation and we present first generalizations to more complex electromagnetic environments in the SI. Entering the realm of current-DFT has furthermore the advantage that usage of the Vignale-Kohn functional [69] provides access to non-adiabatic electron-electron interactions which improves the description of electronic relaxation processes inside the nanoplasmonic particle.

Implementing the radiation-reaction potential into the large-scale TDDFT code GPAW illustrates the high accessibility and seamless integration of our approach into existing TDDFT libraries. In particular, this high accessibility represents a decisive strength, paving the way for a stronger integration of light-matter and open-system features into the popular electronic-structure approaches.

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Figure 4. Absorption spectrum in x-direction for isolated benzene (blue, magenta) and benzene strongly coupled to Al$_{201}$ (black, red, yellow - upper and lower hybrid state P$_{\pm}$ indicated). Dashed(-dotted) lines include emission via the radiation-reaction potential with polarization $e_r = e_z$. The sharp excitation of benzene is only marginally broadened when including emission (magenta, dashed-dotted), i.e., atoms and molecules have comparably long lifetimes. The localized surface-plasmon of the Al$_{201}$, dashed-dotted, i.e., atoms and molecules have comparably long lifetimes. The localized surface-plasmon of the Al$_{201}$ cluster radiates on the other hand substantially stronger (green, dashed). Coupled benzene inherits the short lifetime of the plasmon (red/yellow dashed-dotted) – any excitation is quickly transferred into the plasmon and either internally dephased or subsequently radiated into free-space. Numerical details in_SI
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I. LIGHT-MATTER HAMILTONIAN IN DIPOLE APPROXIMATION

We provide here a short derivation of the dipolar light-matter Hamiltonian. Starting with the free Maxwell-equations, we solve the wave-equation \( \nabla^2 \mathbf{A}(r) - \frac{1}{c^2} \partial_t^2 \mathbf{A}(r) = 0 \) implying the transversality condition of the Coulomb gauge \( \nabla \cdot \mathbf{A}(r) = 0 \). Notice that the Coulomb gauge should be chosen as otherwise the Coulomb interaction takes a different form and thus all electronic structure theory includes the wrong local description. This is a natural limitation for all QED quantization strategies and becomes problematic when quantizing for instance dissipative partially longitudinal modes such as in macroscopic QED. Certainly the ideal situation would be to counteract any gauge-choice for the quantized modes by adjusting the electronic structure calculations but given the extend of electronic structure literature, this is a tedious task. It is therefore essential to only quantize the transverse modes in the following. Only transversal modes can emit into free space and contribute to the far-field radiation while longitudinal components decay quickly [1].

A rectangular box \( V = \prod_{i=1}^{d} L_i \) with perfect conductor boundary condition \( \mathbf{n} \times \mathbf{E}_\perp(r) \propto \mathbf{n} \times \mathbf{A}(r) = 0 \) delivers a set of transverse eigenmodes of the form [2]

\[
\mathbf{S}_{k\lambda}(r) = \sqrt{\frac{2^3}{V}} \begin{pmatrix} \epsilon_{k\lambda}^{(x)} \cos(k_x x) \sin(k_y y) \sin(k_z z) \\ \epsilon_{k\lambda}^{(y)} \sin(k_x x) \cos(k_y y) \sin(k_z z) \\ \epsilon_{k\lambda}^{(z)} \sin(k_x x) \sin(k_y y) \cos(k_z z) \end{pmatrix}
\]

with momentum \( k = (k_x, k_y, k_z)^T \), polarization-index \( \lambda \) for the polarization vector \( \epsilon_{k\lambda} \) and free-field dispersion from the Helmholtz-equation \( \omega(k) = \sqrt{\epsilon(k)} \). The photonic field coordinates are represented by the harmonic oscillator coordinate \( q_{k\lambda} \). Electric and magnetic fields are given by

\[
\mathbf{A}(r, t) = \sum_{k\lambda} \sqrt{\frac{c^2}{\epsilon_0}} q_{k\lambda}(t) \mathbf{S}_{k\lambda}(r)
\]

\[
\mathbf{E}_\perp(r, t) = -\frac{1}{c} \frac{\partial}{\partial t} \mathbf{A}(r, t)
\]

\[
\mathbf{B}(r, t) = \frac{1}{c} \nabla \times \mathbf{A}(r, t)
\]

and construct the transverse electromagnetic field energy

\[
E_\perp = \frac{c_0}{2} \int d^3 r E_\perp(r, t)^2 + c^2 B(r, t)^2.
\]

In the next step, we promote the canonical displacement and momentum coordinates to operators

\[
\hat{q}_k = \left( \frac{\hbar}{2\omega_k} \right)^{1/2} \left( \hat{a}_k^\dagger + \hat{a}_k \right)
\]

\[
\hat{p}_k = \frac{i}{\hbar} \left( \frac{\hbar\omega_k}{2} \right)^{1/2} \left( \hat{a}_k^\dagger - \hat{a}_k \right)
\]

with \( \{\hat{a}_i, \hat{a}_j^\dagger\} = \delta_{ij} \) such that

\[
\hat{\mathbf{A}}(r) = \sum_{k\lambda} \sqrt{\frac{c^2}{\epsilon_0}} \hat{q}_{k\lambda} \mathbf{S}_{k\lambda}(r)
\]

\[
\hat{\mathbf{E}}_\perp(r) = -\sum_{k\lambda} \sqrt{\frac{1}{\epsilon_0}} \hat{p}_{k\lambda} \mathbf{S}_{k\lambda}(r)
\]

\[
\hat{\mathbf{B}}(r) = \sum_{k\lambda} \sqrt{\frac{1}{\epsilon_0}} \hat{q}_{k\lambda} \nabla \times \mathbf{S}_{k\lambda}(r).
\]

The full minimal-coupling light-matter Hamiltonian in Coulomb-gauge reads then (for nuclear and electronic coordinates \( q_i \in \{ \pm eZ_i, -e \} \), \( m_i \in \{ M_n, m_e \} \), \( \bar{p}_i \in \{ -i\hbar \nabla N_{n_i}, -i\hbar \nabla N_{e_i} \} \))

\[
\hat{H}_{lm} = \sum_{i=1}^{N_e+N_n} \frac{1}{2m_i} \left( \bar{p}_i - \frac{q_i}{c} \hat{\mathbf{A}}(r) \right)^2 + \hat{H}_{ph} + \hat{V}_{ext}(r, \mathbf{R}_n)
\]

\[
+ \hat{W}_{ee}(r, r') + \hat{W}_{en}(\mathbf{R}_n, \mathbf{R}_e') + \hat{W}_{ne}(\mathbf{R}_e, \mathbf{R}_n)
\]

\[
\hat{H}_{ph} = \frac{\epsilon_0}{2} \int d^3 r \mathbf{E}_\perp(r, t)^2 + c^2 \hat{\mathbf{B}}(r, t)^2
\]

\[
= \frac{1}{2} \sum_{k\lambda} \sum_{\lambda=1}^2 \hat{p}_{k\lambda}^2 + \omega(k)^2 \hat{q}_{k\lambda}^2.
\]
We assume now that in the coupling-part around the domain over which the matter-system is expanded, the eigenmodes eq. (1) remain constant $S_{k\lambda}(\mathbf{r}) \approx S_{k\lambda}(\mathbf{r}_0)$. Next, we employ the unitary Power-Zienau-Wooley transformation $[1, 3]$ with the total dipole

$$\dot{\mathbf{R}} = -\frac{N^e_{\mathbf{r}}}{} + \frac{N^b_{\mathbf{r}}}{} + \sum_{\mu=1}^n \mathbf{e}_\mu \mathbf{C}_\mu^a. \]$$

This transformation is nothing else than a momentum-translation which will transfer the coupling from momentum-fluctuations into displacement-fluctuations $[4]$. By doing so, we remove the photonic part from the covariant momentum

\[ \left( \dot{p}_i/m - q_i/c \mathbf{A}(\mathbf{r}_{\text{matter}}) \right) \rightarrow \dot{p}_i/m \] 

and introduce coupling in the previously pure photonic Hamiltonian $\hat{H}_{\text{ph}} \rightarrow \hat{H}_{\text{ep}}$. Performing a Fourier-transformation in the photonic coordinates $\hat{p}_{k\lambda} \rightarrow -\omega(\mathbf{k}) \hat{q}_{k\lambda}$, $\hat{q}_{k\lambda} \rightarrow 1/\omega(\mathbf{k}) \hat{p}_{k\lambda}$ leads finally to

\[ \dot{\mathbf{R}} = \dot{\mathbf{T}} + \dot{\mathbf{V}} + \dot{\mathbf{W}} + \dot{\mathbf{H}}_{\text{ep}} \] 

\[ \dot{\mathbf{H}}_{\text{ep}} = \frac{1}{2} \sum_{\lambda=1}^{2} \sum_{\mathbf{k}} (\hat{S}_{k\lambda}^2 + \omega(\mathbf{k})^2 \left[ \hat{q}_{k\lambda} - \frac{\lambda_{k\lambda}(\mathbf{r}_0)}{\omega(\mathbf{k})} \cdot \hat{\mathbf{R}} \right] ) \] 

with

\[ \lambda_{k\lambda}(\mathbf{r}) = \frac{1}{\sqrt{\varepsilon_0}} S_{k\lambda}(\mathbf{r}) \] 

Here $\mathbf{r}_0$ should be ideally the center of charge of the whole matter system. The PZW transformation also changes the field quantities, i.e., the meaning of $\hat{a}$ changes. The electric field is no longer a purely photonic quantity but includes the transversal polarization

\[ \varepsilon_0 \hat{\mathbf{E}}_\perp = (\hat{\mathbf{D}}_\perp - \hat{\mathbf{P}}_\perp) \] 

\[ \hat{\mathbf{P}}_\perp = \sum_{k\lambda} \hat{q}_{k\lambda} (\lambda_{k\lambda} \cdot \hat{\mathbf{R}}) \] 

Consequently, $\mathbf{E}$ is no longer the conjugated momentum to $\mathbf{A}$ but instead the displacement field steps into this role

\[ \hat{\mathbf{D}}_\perp(\mathbf{r}) = \varepsilon_0 \sum_{k\lambda} \lambda_{k\lambda}(\mathbf{r}_0) \omega(\mathbf{k}) \hat{q}_{k\lambda} \] 

We will disregard the coupling to the nuclei for the TDDFT derivation.

II. RADIATION-REACTION FOR ARBITRARY PHOTONIC ENVIRONMENTS

One-dimensional emission, as present in waveguides or idealized cavities, is surely only one out of many relevant realizations of photonic environments. In general, the classical electromagnetic mode structure inside a given (lossy) resonator, which may be transversal and longitudinal, can be deduced by solving the Helmholtz equation

\[ \nabla \times \left( \frac{1}{\mu_0(\mathbf{r},\omega)} \nabla \times -\omega^2 \mu_0 \varepsilon_0(\mathbf{r},\omega) \right) G(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') \] 

providing as solution the dyadic Greens tensor. Certainly, this path generalizes also radiation-reaction as known for perturbative treatments $[5]$. It is important here to recall the observations of the previous section. In general, the dipolar coupling appears between displacement field and dipole $\mathbf{r}(\mathbf{t}) = \mathbf{e} \cdot \mathbf{D}_\perp(\mathbf{t})/\varepsilon_0$ with $\mathbf{D}_\perp = \varepsilon_0 \mathbf{E}_\perp + \mathbf{P}_\perp \approx \varepsilon_0 \varepsilon_0(\mathbf{r},\omega) \mathbf{E}_\perp$, i.e., the coupling fields are not the same as the radiated fields. We ignored this aspect in the main text for brevity as it does not influence the presented free-space emission results. Quantization of quasi-normalmodes has been explored by macroscopic QED and associated approaches $[6, 7]$. The target of most of these techniques is to translate the complex mode-structure around sizable plasmonic systems into easy to handle (quasi) normal-modes which allows the usage of common quantum optical techniques to describe e.g. emission via the plasmonic modes. The number of those normal-modes will be considerable and commonly only perturbative usage along the lines of Wigner-Weisskopf theory is employed. As already briefly discussed in the main document and above, radiation-reaction can be adjusted accordingly to account for a more complex mode-structure. This will likely prevent analytical integrations similar to the free-space case illustrated in the main text and demand the numerical expression for the Greens tensor. The latter implies the use of widely available Maxwell solvers and is thus not a conceptual problem but will merely lead to a more involved expression for the radiation-reaction potential. The description of small plasmonic systems can be done most consistently by simply describing also the plasmonic system using TDDFT on equal footing with any molecular particle close to it $[8]$. Obvious computational limitations will prevent us from pushing this ‘single-system’ description to arbitrary system-size which motivates mixed quantum-classical descriptions such as combining Mie-theory and TDDFT via the radiation-reaction potential. Consider hereby the disclaimer at the start of this document regarding the consistency of gauges, i.e., electronic structure theory is usually defined in the Coulomb gauge and the radiation-reaction forces should be consistent with this choice.

A. Single-dimensional time-dependent radiation-reaction potential via dyadic Greens function

We start from the expressions $v_\mathbf{e}(\mathbf{r},\mathbf{t}) = -\mathbf{e} \cdot \mathbf{E}_\mathbf{r}(\mathbf{t})$ and $\mathbf{E}_\mathbf{r}(\mathbf{r},\omega) = i \mu_0 \omega \int_{\mathbf{r}} d\mathbf{r}' G(\mathbf{r}, \mathbf{r}', \omega) \cdot (\mathbf{e} \cdot (\mathbf{e} \cdot \mathbf{r}'))$ using the Greens function as defined by the spectral theorem

\[ G(\mathbf{x}, \mathbf{x}', \omega) = \sum_{k\lambda} \frac{S(x)S'(x')}{\varepsilon_0(\omega/\varepsilon_0)} e_\lambda^e e_{\lambda'}^e \] 

with $S(x) = \sqrt{1/V(\cos(2\pi x/L) + \sin(2\pi x/L))}$. The electric field can be grouped into two contributions that in frequency space

\[ \frac{1}{k^2 - \omega^2/c^2} \] 

and $i\omega j(x)$. In order to obtain the time-dependent radiation-reaction field, we make use of the convolution theorem for
the inverse Fourier-transformation \( F^{-1}(E_r(r, \omega)) \propto F^{-1}\left(\frac{i\omega j(r, \omega)}{k^2 - \omega^2/c^2}\right) = F^{-1}\left(\frac{1}{k^2 - \omega^2/c^2}\right) \ast F^{-1}(i\omega j(r, \omega)) \).

The last part is trivially obtained as \( F^{-1}(i\omega j(r, \omega)) = -\partial_t j(r, t) \). The first part is formally solved by complex integration around the two poles at \( k \pm \omega/c \)

\[
\int \frac{dk}{2\pi} e^{ikt} \frac{1}{(k - \omega/c)(k + \omega/c)} = e^{\frac{ic}{2k}} (e^{ikt} - e^{-ikt}), \quad t > 0; \quad \text{and 0 otherwise.}
\]

Let us absorb the causality \( t > 0 \) into the Heaviside function \( \theta(t) \) and combine the exponentials into the sine function. The convolution is then

\[
\int_{-\infty}^{\infty} dt' (t - t') c^2 \sin(c(k(t - t')))(-1)\partial_t j(r, t')
\]

and by partial integration (assuming \( j(r, -\infty) = 0 \))

\[
\int_{-\infty}^{t} dt' c^2 \cos(c(k(t - t')))j(r, t') \quad \text{The negative infinity is hereby limited by the initial conditions (see e.g. the following section). Our radiated field is then given by (free-space} \ c = \omega, \ \mu_0 = 1/(\varepsilon_0 c^2)\)

\[
E_r(x, t) = e_e \int_{V} dr' \sum_k \frac{1}{\varepsilon_0} S(x)S(x') \int_{-\infty}^{t} dt' \cos(c(k(t - t')))j_e \cdot (-e)_j(r', t') \quad \text{and the radiation-reaction potential at the center of charge} \ x = x_o = 0 \text{ follows immediately} \ v_{rr}(r, t) = e_e \cdot E_{rr}(t) = e_e \sum_k \frac{1}{\varepsilon_0} \int_{V} dr' \cos(c(k(t - t'))) \int_{V} dr' e_e \cdot (-e)_j(r', t') \quad \text{With the help of the continuity equation} \ R = \int dr - e_j(r, t) \text{ we obtain the compact form}

\[
v_{rr}(r, t) = e_e \sum_k \frac{1}{\varepsilon_0} \int_{-\infty}^{t} dt' \cos(c(k(t - t')))j_e \cdot R(t').
\]

As we will see in the following section, this form is identical to the Maxwell/Ehrenfest potential in QEDFT describing the interaction between a set of harmonic oscillators and the electronic system. Turning the sum over modes into an explicit integration results ultimately in the radiation-reaction potential eq. (12) as shown in the main text.

### B. Single-dimensional free-space emission as analytic limit from QEDFT

Starting with the seminal work by Tokatly [9], it is possible to explicitly derive a local potential for the Ehrenfest interaction between a set of photonic modes and the electronic system in the long-wave approximation

\[
v_{M}(r, t) = \sum_{k\lambda} \lambda_{k\lambda} \cdot e_r \left[\omega_{k\lambda} q_{k\lambda}(t) - \lambda_{k\lambda} \cdot R(t)\right].
\]

The mode-resolved Maxwell equations describing those eigenmodes \( \partial_{t}^{2} q_{k\lambda}(t) + \omega_{k\lambda}^{2} q_{k\lambda}(t) = \omega_{k\lambda} \lambda_{k\lambda} \cdot R(t) \) can be solved with the help of the classical Greens function \( \sin(\omega_{k\lambda}(t - t'))/\omega_{k\lambda} \).

Let us assume that at \( t = t_0 \) no time-dynamic was present (in the interacting and the Kohn-Sham system), i.e., \( \partial_{t} q_{k\lambda}(t_0) = \partial_{t} R(t_0) = 0 \).

Then \( q_{k\lambda}(t_0) = \lambda_{k\lambda} \cdot R(t_0)/\omega_{k\lambda} \) and we obtain after partial integration

\[
v_{M}(r, t) = \sum_{k\lambda} \lambda_{k\lambda} \cdot e_r \int_{t_0}^{t} dt' \cos(\omega_{k\lambda}(t - t')) \lambda_{k\lambda} \cdot R(t').
\]

With \( S(0) = \sqrt{1/V}, \ k_x = 2\pi n_x/L_x, \ n_x \in \mathbb{Z}, \sum_{k\lambda} = \frac{L_x}{2\pi} \int_{-\infty}^{\infty} dk, \) performing the explicit integration and we obtain with \( \frac{\sin(\omega_{k\lambda}(t - t'))}{\omega_{k\lambda}} \rightarrow \delta(t - t') \) the radiation-reaction potential describing one-dimensional emission within TDDFT as defined in the main text

\[
v_{r\perp}^{1D}(r, t) = \frac{4\pi \hbar \alpha}{c^2} A^{-1} e_e \cdot R(t) \quad e_e \cdot (-e_r). \quad (12)
\]

We will focus in the following on other analytic limits that provide a very intuitive understanding of the radiation-reaction potential. First, the presence of two mirrors will modulate the free-space emission which illustrates nicely the conceptual step that has to be performed in order to describe more general systems. Finally, we extend our derivations to full three-dimensional emission which allows us to connect radiation-reaction to its historical counterpart, the Abraham-Lorentz model.

### C. Edge-emission from a cavity

Let us assume we describe a cavity with confined modes in the \( z \) direction while we allow for free-space emission in \( x \) direction and align the polarization of all modes in the \( z \) direction while we allow for free-space emission which allows us to connect the radiation-reaction potential to its historical concept, the Abraham-Lorentz model.

Focusing on the first term

\[
\sum_{k \perp z} = \frac{L_x}{2\pi} \int_{-\infty}^{\infty} dk \int_{0}^{\infty} dz \int_{-\infty}^{\infty} dk d\phi_m
\]

where we introduced polar coordinates in the \( x-z \) plane and assumed that we only treat k’s which are very close to the \( x \)-axis. Setting \( x = 0 \) without loss of generality, we then have

\[
\sum_{k \perp z} \lambda_k \lambda_k \cos(\omega_k(t - t'))
\]

\[
= \frac{L_x L_z}{\varepsilon_0 V \pi^2} \int_{0}^{\phi_m} dk d\phi_m \int_{-\phi_m}^{\phi_m} \sin^2(k \sin(\phi) z_0) \cos(\omega_k(t - t'))
\]
which for small $\phi$ can be expanded as

$$\phi_m = \frac{1}{\varepsilon_0 \pi^2 L_y} \int_0^\infty dk \int_{-\phi_m}^{\phi_m} \sin^2(k\phi z_0) \cos(\omega_k(t - t'))$$

Performing the integration over $\phi$ leads to

$$\frac{1}{\varepsilon_0 \pi^2 L_y} \int_0^\infty dk k \sin(\omega_k(t - t')) \left( \phi_m - \frac{\sin(2\phi_m k z_0)}{2k z_0} \right)$$

Next, we have to find an expression for $\phi_m$. The angle $\phi = \arctan(k_z/k_x)$ is in our case approximately $\approx k_z/k_x$ and we know that for $\omega \to \infty$, $\omega = ck \approx ck_x$. The limit we are therefore about to perform suggests that

$$\phi_m \approx \frac{k_z}{k_x} \approx \frac{c n_z}{\omega L_z}$$

Only for small $k_z/k_x$ and thus $n_z \in \mathbb{N} \approx 1$ does our approximation make sense and we therefore set $n_z = 1$ from here on. Since $\phi_m \propto \frac{1}{\varepsilon_0 \pi^2 L_y}$, we can now obtain an analytic solution for the $dk = \frac{dz}{c}$, $k = \frac{z}{c}$ integral

$$\frac{1}{\varepsilon_0 \pi^2 L_y L_z} \left(1 - \frac{\sin(2\pi z_0/L_z)}{2\pi z_0/L_z} \right) \delta(t - t')$$

The sine expression tends for $z_0 \to 0$ towards $1$, i.e., next to the mirror we do not have any available mode-volume and the emission is quenched (destructive Purcell). However, if we place our system in the center of the cavity $z_0 = L_z/2$, we obtain $\sin(\pi) = 0$ and thus 'standard' emission along the $x$ direction. Notice that $z_0 = \frac{3L_z}{2}$, $\sin(3\pi/2)/(3\pi/2) \approx -0.2122$ is small but negative, i.e., weak amplifications can appear depending on the position along $z_0$. This behavior resembles the typical dipole emission next to a perfect conductor plate [10].

While this provides the expected emission-behavior of a dipole next to a plate, we have two plates, the second at distance $z_0 = L_z$. Taking this into account, a symmetrized multiplicative form should be used similar to

$$\left(1 - \frac{\sin(2\pi z_0/L_z)}{2\pi z_0/L_z} \right) \left(1 - \frac{\sin(2\pi(1 - z_0/L_z))}{2\pi(1 - z_0/L_z)} \right)$$

Under those conditions and considering $1/\varepsilon_0 c = 4\pi \hbar c/e^2$, we obtain the radiation-reaction potential for perpendicular emission from a cavity (edge emission from a cavity with polarization along $y$) as

$$v_{rr}^{\phi \phi e}(rt') = v_{rr}^{1D}(rt) \left(1 - \frac{\sin(2\pi z_0/L_z)}{2\pi z_0/L_z} \right) \left(1 - \frac{\sin(2\pi(1 - z_0/L_z))}{2\pi(1 - z_0/L_z)} \right)$$

which is only slightly modified with respect to the pure free-space emission $v_{rr}^{1D}$.

### D. Three-dimensional free-space emission and its connection to the Abraham-Lorentz model

With increasing dimensionality of the photonic environment, the density of photonic modes increases $\mathcal{O}(r_{ph}^{d-1})$. This rapid increase in mode density renders a direct integration problematic unless we explicitly assume some form of Markovian approximation. The generic form of the radiation-reaction emission using the dyadic greens tensor, which for the emission of spherical waves is given by $G(r, r', \omega) = 1/(4\pi) e^{ik|r-r'|/|r-r'|}$, provides a convenient approach to this problem. In the following we will however stay consistent with the line of thought chosen in the previous sections. The 3D free-space emission can be described by a periodic box with couple $\lambda_{k\lambda}(r_0 = 0) = \sqrt{\frac{1}{\varepsilon_0 e^2}}\kappa_{k\lambda}$ at its center where $k = \frac{2\pi}{\lambda} n$, $n \in \mathbb{Z}$.

The corresponding potential is then given by

$$v_{3D}(rt) = (-er_{\mu}) \int_{t_0}^t \int_{t_0}^t \int_{k\lambda}^\infty d\kappa \int_{\lambda_{k\lambda}}^\infty d\lambda \int_0^{2\pi} 0 \sin(\theta) 0 \cos(\omega_{k\lambda}(t - t'))$$

with

$$\kappa \lambda_{k\lambda} \cdot \lambda_{k\lambda} = (r_x \sin \theta \cos \phi + r_y \sin \theta \sin \phi + r_z \cos \theta)$$

$$\int_0^\pi 0 \sin(\theta) 0 \cos(\omega_{k\lambda}(t - t'))$$

such that

$$v_{3D}(rt) = \frac{1}{3\varepsilon_0 \pi^2} \int_{t_0}^t \int_{t_0}^t \int_{k_{\lambda}}^\infty \kappa \lambda d\kappa d\lambda \int_0^{2\pi} 0 \sin(\theta) 0 \cos(\omega_{k\lambda}(t - t'))$$

where $\omega_{k\lambda} = c|k| = ck$. We are left now with an expression that will diverge if we would naively integrate up to infinite frequencies. A common procedure is to introduce a physical cut-off, e.g. the pair-creation energy, which leads in the routinely followed paths to the physical mass of the electron (as we have already seen in the main text). We follow here two alternative ways that avoid the introduction of a cut-off, presenting first a more quantum-optical ansatz following the Markovian approximation, followed by the connection to the classical Abraham-Lorentz model.

Let us assume the molecular dipole oscillates 'almost' freely, thus the effect of the free-field is vanishingly small and the molecule behaves close to harmonic $(\partial^2 \gamma + \omega_n^2) R(t') = 0$. Then, the dipole propagating from $t$ to $t'$ would be $R(t') = R(t) \cos(\omega_n(t' - t)) + \dot{R}(t) \sin(\omega_n(t' - t))/\omega_n$. We now place this Markovian-like ansatz for the dipole into the original potential, use
\[
\cos(x)\cos(y) = \frac{1}{2}(\cos(x+y) + \cos(x-y)), \sin(x)\cos(y) = \frac{1}{2}(\sin(x+y) - \sin(x-y)) \text{ and resolve the integral } \int dt' \text{ to obtain}
\]
\[
= \frac{1}{3\varepsilon_0 \pi^2} \int_{t_0}^{t} dt' \int_{0}^{k_{\max}} dk k^2 (-er) \left[ \tilde{R}(t) \sin[(\omega_n - \omega_k)(t-t_0)] + \text{counter-RWA} \right] - R(t) \frac{\omega_n \cos[(\omega_n - \omega_k)(t-t_0)]}{[\omega_n - \omega_k]} + \text{counter-RWA}. \]

We added here a factor 2 to the expression to correctly recover the Wigner-Weisskopf rate in the following, the simplified Markovian-like ansatz is likely to imply this correction. The counter-rotating terms can be neglected and in the limit \( t \to \infty \) one obtains \( \sin[(\omega_n - \omega_k)(t-t_0)] \to \pi \delta(\omega_n - \omega_k) \) as well as \( \cos[(\omega_n - \omega_k)(t-t_0)] \to \omega_n \) which remains oscillating and we thus disregard it here. With \( dk = \frac{d\omega}{c}, \) \( k = \frac{\omega}{c} \) we obtain
\[
\frac{\tilde{R}(t)}{3\varepsilon_0 \pi c^3} \int_{0}^{k_{\max}} d\omega \omega^2 (-er) \delta(\omega_n - \omega_k) \]
\[
\frac{\omega_n^2}{3\varepsilon_0 \pi c^3} (-er) \cdot \tilde{R}(t) \]
the 3D isotropic free-space ‘radiation-reaction’ emission potential. Following the previous linear response derivation, the natural linewidth
\[
\Gamma_{3D}^{rr} = \frac{\omega_n^3 |\Phi_n|^2}{3\hbar \varepsilon_0 \pi c^3}
\]
is identical to the free-space Wigner-Weisskopf rate.

Alternatively, we could identify \( k^2 \) to be represented by time-derivatives. Following partial integration
\[
\int_{t_0}^{t} dt' \left[ -\frac{k}{c} \partial_t^2 \cos(ck(t-t')) \tilde{R}(t') \right]
\]
\[
= \frac{k}{c} \sin(ck(t-t')) \tilde{R}(t') |_{t'=t_0} - \int_{t_0}^{t} dt' \left[ -\frac{1}{c^2} \partial_t \cos(ck(t-t')) \tilde{R}(t') \right]
\]
\[
= \frac{k}{c} \sin(ck(t-t')) \tilde{R}(t') |_{t'=t_0} - \int_{t_0}^{t} dt' \left[ -\frac{1}{c^2} \cos(ck(t-t')) \tilde{R}(t') |_{t'=t_0} \right]
\]
identifying the boundary terms as 0 or divergent
\[
\int dk \left[ -\frac{k}{c} \sin(ck(t-t')) + \tilde{R}(t') \right] \frac{\tilde{R}(t_0)}{c^2} \cos(ck(t-t_0)) \frac{\tilde{R}(t_0)}{c^2} \cos(ck(t-t_0)) \right]
\]
\[
= \frac{\tilde{R}(t_0)}{c^2} \sin(ck_{\max}(t-t_0)) \frac{\tilde{R}(t_0)}{c^2} \cos(ck_{\max}(t-t_0)) \right] \to -\infty \to -\infty \to -\frac{\pi}{c^3} \to 0
\]
we are left with
\[
\int dk \cos(ck(t-t')) = \frac{\sin(ck(t-t'))}{c(t-t')} \to \frac{\pi}{c} \delta(t-t').
\]

We then obtain the simplified Abraham-Lorentz radiation-reaction potential
\[
v_{rr}^{AL}(rt) = -\frac{1}{3\pi \varepsilon_0 c^2} (-er) \cdot \tilde{R}(t).
\]

Apparent from the third time-derivative, the dynamics and the linear response of a system driven by such a local potential becomes non-trivial. Our radiation-reaction potential is now structurally very similar to the Abraham-Lorentz force
\[
F_{AL} = -\frac{2e^2}{3 \cdot 4\pi \varepsilon_0 c^3} \dddot{x}(t).
\]

Such a radiation-reaction jerk \( \dddot{x} \), and the associated classical Newton equations, lead to non-causal pre-acceleration features and runaway solutions. The non-causal features arising from the Abraham-Lorentz model troubled famous physicists for decades and have only been resolved by quantum electrodynamics.

Using an alternative expression for the second derivative of the density in terms of the forces exerted by the Kohn-Sham stress-tensor \( T_{ik}(r) = \frac{1}{2} [\partial_i \Psi^\dagger(r) \partial_k \Psi(r) + \partial_k \Psi^\dagger(r) \partial_i \Psi(r) - \frac{1}{2} \partial_i \partial_k \{ \Psi^\dagger(r) \Psi(r) \} ] \), \[11\]
\[
\partial^2 \rho(r) = \nabla \cdot [\rho(r) \nabla v_s(r)] + \sum_{i,k} \partial_i \partial_k \dot{T}_{ik}(r)
\]
we could avoid the nonphysical third derivative. The dependence of \( \partial^2 \rho \) on the radiation-reaction contribution in \( v_s \) includes now the self-consistency that was previously encoded in the higher derivative. In a first intuitive step, one could discard the self-consistency of the radiation-reaction potential, i.e., \( v_s = v_{ext} + v_{Hxc} \), but the detailed analysis necessary to judge the quality (including the question of bijectivity between potential and density) of the suggested approximations extends beyond the scope of this work.

Let us finally recall out that the here illustrated approach can be extended beyond the common long wavelength approximation. Furthermore, by utilizing for instance the recently suggested photon-free framework for QEDFT [12], quantum effects of the photonic system can be intuitively included.

### III. NUMERICAL DETAILS

Inputs and code are available upon reasonable request. In order to not interfere with the linear response kick, the radiation-reaction potential has been switched on always shortly after the kick (hydrogen runs at \( t=2 \) a.u., GPAW at \( t=5 \) a.u.). Fig. 1 and 2 utilized a time-stepping of \( \Delta t = 10^{-2} \) a.u., a real-space grid with 301 grid-points and equidistant spacing \( \Delta x = 0.1 \) a.u.. For Fig. 2, the system is initialized in the ground state, perturbed by a linear response kick \( v_{kick}(x,t) = -10^{-6} x / \pi [(t-1)^2 + 10^{-4}] \) (in a.u.) and propagated for 4000 a.u. of time. We extended...
the propagation time by a factor of 5 for the EIT curve ($g/\hbar \omega_c = 0.01$) in order to improve the frequency resolution and clearly resolve the dip to 0 absorption.

A. Perturbative to non-perturbative emission limit

In order to numerically differentiate between the physical lifetime and the artificial width of excitations in real-time TDDFT due to finite propagation time, the damping of the dipole has to be considerable. For weakly confined electromagnetic environments, the lifetime of small isolated atomic and molecular systems is substantially longer than typical simulation times in TDDFT and it is therefore advisable to deduce the linewidth by selecting a larger $A^{-1}$ than physically available and extrapolating towards the correct value [13] or to alternatively follow the Casida linear-response approach. As we have seen in the main text, some systems involving plasmonic coupling or strongly confined waveguides can strongly enhance the emission characteristics and shorten the lifetimes.

Figure 1 illustrates the cross-over between purely perturbative emission which follows Wigner-Weisskopf theory (gray dashed inset) and the non-perturbative limit in which the rate remains below the perturbative extrapolation. The good agreement in the perturbative limit is consistent with the computationally more involved explicit sampling QEDFT Casida description utilized by Flick et al. [14]. As long as different excitations are clearly separated, the perturbative description provide an adequate estimate for the emission rate. Clearly hydrogen is hereby a pathological example due to the simplicity of the electronic structure and even when the linewidth deviates from the well defined Lorentzian, Wigner-Weisskopf remains a good estimate.

B. Cross section

The in fig. 2 provided photo-absorption cross-section $\sigma(\omega) = 4\pi \omega^3 \alpha(\omega)$ (single dimension $\alpha(\omega) = \alpha_{xx}(\omega)$) has been calculated straight away from the definition $\alpha_{xx}(\omega) = R_{xx}(\omega)/E_x(\omega)$. No additional window functions have been used in the FFT.

C. Na chain

Fig. 4 has been obtained by perturbing the ground state by a linear response kick of strength $10^{-5}$ (ASE units). The ground state and basis has been obtained in a simulation box of dimensions 96 A x 12 A x 12 A. The LDA functional, the standard Poisson solver, removing the momenta 1, 3 and 5, as well as a grid-spacing of 0.3 A and 12 (6 for Na$_2$) bands have been used. We propagate 3000 (8000 for Na$_2$) steps with a time-step of 10 A/\sqrt{\mu/eV} and the double zeta polarized LCAO basis. The spectral function was folded with a sharp Lorentzian of width $10^{-3}$ to smoothen the results, the observed linewidth is a natural feature of the damped propagation due to the radiation-reaction forces and is not introduced by this smoothening procedure but artificial oscillations appear due to sinc contamination. Fig. 2 shows the direct FFT of the dipole moment.

D. Al$_{201}$C$_6$H$_6$

We use the structures and parameters published in [8] (structure name Al201+b1,0). The spectrum has been smoothed via convolution with a sharp Lorentzian of width 0.01 eV in order to clearly illustrate the considerable effect of the plasmon on the lifetime of benzene.
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