Dressed-Orbital Approach to Cavity Quantum Electrodynamics and Beyond

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We present a novel representation of coupled matter-photon systems that allows the application of *any* many-body method developed for purely fermionic systems. We do so by rewriting the original coupled light-matter problem in a higher-dimensional configuration space and then use photon-dressed orbitals as a basis to expand the thus "fermionized" coupled system. As an application we present a dressed time-dependent density-functional theory (TDDFT) approach. The resulting dressed Kohn-Sham (KS) scheme allows for straightforward non-adiabatic approximations to the unknown exchange-correlation potential that explicitly includes correlations. We highlight this for simple model systems placed inside a high-Q optical cavity and show also results for novel types of observables such as photon-field fluctuations. We finally highlight how the dressed-orbital approach goes beyond the context of cavity quantum electrodynamics and can be applied to, e.g., van-der-Waals problems.

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In the last decade experiments at the interface between chemistry, material science and quantum optics have uncovered situations in which the strong interplay between the quantized electromagnetic field and the matter degrees of freedom lead to interesting physical phenomena and novel states of matter [1–4]. For instance, by strongly coupling molecules to the vacuum field of an optical cavity chemical reactions can be modified [5, 6], strong exciton-photon coupling in light-harvesting complexes is expected to modify energy transfer [7] or the coupling to quantum matter can lead to attractive photons [8]. Experimental results show that the emergence of hybrid light-matter states, so-called polaritonic states that strongly mix matter and photon degrees of freedom, is the reason for these interesting phenomena. While the theoretical understanding of these effects are mostly based on simplified Dicke-type models (several few-level systems coupled to one mode) [9], an accurate and unbiased description of the physical situation calls for calculations of the coupled matter-photon systems from *first principles* [3, 4, 10, 11]. An appealing such first-principles method is an extension of density-functional theory to coupled matter-photon systems [12–15], which is called quantum-electrodynamical density-functional theory (QEDFT). By reformulating the fully coupled fermion-boson problem in a formally exact quantum-fluid description, where the charge current is coupled in a non-linear way to the Maxwell field, it avoids a solution in terms of the usually infeasible wave function. The major drawback of such density-functional approaches are that the internal force terms of the quantum fluids are only known implicitly (in terms of the wave function) and approximations have to be used in general. The most successful approximation strategy is the Kohn-Sham (KS) scheme, where the local-force expressions of a non-interacting auxiliary system is used as a starting point to model the fully interacting case [16, 17]. First calculations for real molecules coupled to photons in and out of equilibrium [18, 19] show the potential of first-principle calculations of coupled matter-photon systems. However, approximations based on the standard KS scheme are hard to improve towards strongly-coupled systems, which in the context of coupled matter-photon systems promise interesting physical effects [20]. Alternative approximation schemes either rely on a different auxiliary system, such as the strictly-correlated electron system [21] or go beyond the single Slater-determinant Ansatz [22].

In this work we provide a completely different route to describe matter-photon systems from first-principles by reformulating the physical problem in a space with auxiliary extra dimensions. This higher-dimensional reformulation reduces to the standard formulation in physical space in a straightforward manner, providing us with an "holographic" perspective of the original problem that allows us to work with explicitly-correlated/dressed higher-dimensional orbitals. Since this leads to a "fermionization" of the coupled fermion-boson system it allows us to employ *any* fermionic many-body method developed, such as, e.g., Greens' function techniques [23, 24] or density-functional methods. Here we exemplify the possibilities of this approach by applying it in the context of TDDFT and find that already the simplest approximations are non-adiabatic from the start and include explicit correlations that can otherwise only be captured by advanced functionals for the standard KS scheme.

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We illustrate this dressed KS construction for the case of an electronic quantum system coupled to the photons of an optical cavity. We show how simple approximations in terms of dressed (mixed matter-photon) orbitals captures the right Rabi-oscillation induced by the photon-matter coupling for a Rabi model. We further show how it captures the non-adiabatic dynamics of a "bare" model helium that is brought inside an optical cavity (spontaneous emission), and how the dressed KS approach allows to also determine novel types of observables such as the photon field fluctuations. Finally we highlight that this scheme can be used also in other physical situations to capture strong correlations, e.g., for dispersion interactions.

Let us consider the case of a general electronic system with frozen ions inside an optical cavity (the extension to include the nuclei as quantum particles is straightforward [3, 11]). As the spatial extension of the matter system is small compared to the wave length of the cavity modes, we can treat the matter-photon coupling in dipole approximation (atomic units are used throughout) [25, 26]. In this case the interacting Hamiltonian reads

$$\begin{aligned}\hat{H}(t) = &\sum_{k=1}^{N}\left[-\tfrac{1}{2}\nabla^2_{\mathbf{r}_k} + v(\mathbf{r}_k,t)\right] + \tfrac{1}{2}\sum_{k\neq l} w(\mathbf{r}_k,\mathbf{r}_l) \qquad (1)\\ &+\sum_{\alpha=1}^{M}\left[-\tfrac{1}{2}\tfrac{\partial^2}{\partial p_\alpha^2} + \tfrac{1}{2}\Big(\omega_\alpha p_\alpha - \boldsymbol{\lambda}_\alpha\cdot\sum_{k=1}^{N}\mathbf{r}_k\Big)^2 + \tfrac{j_\alpha(t)}{\omega_\alpha}p_\alpha\right],\end{aligned}$$

where the first line corresponds to the usual many-body Hamiltonian $\hat{T}+\hat{V}(t)+\hat{W}$ describing the uncoupled matter system of N electrons interacting via the Coulomb potential $w(\mathbf{r},\mathbf{r}')$ and moving in an external time dependent potential $v(\mathbf{r},t)$. The second line describes M photon modes with frequency ω_α and polarization vectors $\boldsymbol{\lambda}_\alpha$ coupled to the total dipole of the electronic system. Furthermore, the photon modes are allowed to couple to an external source $j_\alpha(t)$ that corresponds to the time-derivative of a mode resolved external current [13, 26].

We first introduce auxiliary extra dimensions $(p_{\alpha,2},...,p_{\alpha,N})$ for each mode, and then consider the extended Hamiltonian $\hat{H}'(t)=\hat{H}(t)+\hat{H}_{\text{aux}}$, with $\hat{H}_{\text{aux}} = \sum_{\alpha=1}^{M}\hat{H}_{\text{aux},\alpha}$ and $\hat{H}_{\text{aux},\alpha} = \sum_{k=2}^{N}\left(-\frac{1}{2}\frac{\partial^2}{\partial p^2_{\alpha,k}} + \frac{\omega_\alpha^2}{2}p^2_{\alpha,k}\right)$. We then change coordinates to $(q_{\alpha,1},...,q_{\alpha,N})$ for each mode, where $(q_{\alpha,1},...,q_{\alpha,N})$ is any coordinate set related to $(p_\alpha,p_{\alpha,2},...,p_{\alpha,N})$ by an orthogonal transformation (the specific choice does not matter) that further allows us to specify the photon-displacement coordinates p_α as

$$p_\alpha = \tfrac{1}{\sqrt{N}}\left(q_{\alpha,1}+...+q_{\alpha,N}\right). \qquad (2)$$

This is the inverse of a center-of-mass coordinate transformation [27] (see Supplemental Material Sec. I for an explicit example). Introducing a $(3+M)$-dimensional dressed vector of space and auxiliary photon coordinates $\mathbf{z}=(\mathbf{r},q_1,...,q_M)$ we can then rewrite $\hat{H}'(t)$ as

$$\begin{aligned}\hat{H}'(t) &= \sum_{k=1}^{N}\left[-\tfrac{1}{2}\nabla^2_{\mathbf{z}_k} + v'(\mathbf{z}_k,t)\right] + \tfrac{1}{2}\sum_{k\neq l} w'(\mathbf{z}_k,\mathbf{z}_l) \qquad (3)\\ &= \hat{T}' + \hat{V}'(t) + \hat{W}',\end{aligned}$$

where $v'(\mathbf{z},t)=v(\mathbf{r},t)+\sum_{\alpha=1}^{M}[\frac{1}{2}\omega_\alpha^2 q_\alpha^2 - \frac{\omega_\alpha}{\sqrt{N}}q_\alpha(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}) + \frac{1}{2}(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r})^2 + \frac{j_\alpha(t)q_\alpha}{\sqrt{N}\omega_\alpha}]$ and $w'(\mathbf{z},\mathbf{z}') = w(\mathbf{r},\mathbf{r}')+\sum_{\alpha=1}^{M}[-\frac{\omega_\alpha}{\sqrt{N}}q_\alpha(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}') - \frac{\omega_\alpha}{\sqrt{N}}q'_\alpha(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}) + (\boldsymbol{\lambda}_\alpha\cdot\mathbf{r})(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}')]$. Here we used that for an orthogonal transformation $\sum_{k=1}^{N}\frac{\partial^2}{\partial p^2_{\alpha,k}} = \sum_{k=1}^{N}\frac{\partial^2}{\partial q^2_{\alpha,k}}$ and $\frac{\omega_\alpha^2}{2}(p_\alpha^2+p_{\alpha,2}^2+...+p_{\alpha,N}^2) = \frac{\omega_\alpha^2}{2}(q_{\alpha,1}^2+...+q_{\alpha,N}^2)$. Now, if the normalized $\Psi(t)$ solves the physical time-dependent Schrödinger equation (TDSE) $\mathrm{i}\frac{\partial}{\partial t}\Psi(t) = \hat{H}(t)\Psi(t)$, then

$$\begin{aligned}&\Psi'(\mathbf{z}_1\sigma_1,...,\mathbf{z}_N\sigma_N,t) =\\ &\Psi(\mathbf{r}_1\sigma_1,...,\mathbf{r}_N\sigma_N,p_1,...,p_M,t)\chi(p_{1,2},...,p_{M,N},t),\end{aligned}$$

is the normalized solution of the extended TDSE $\mathrm{i}\frac{\partial}{\partial t}\Psi'(t) = \hat{H}'(t)\Psi'(t)$ with a $\chi(t)$ that evolves from an arbitrary, normalized initial state χ_0 under $\hat{H}_{\text{aux}}$. Since $\Psi(t)$ depends only on the coordinates defined in Eq. (2), it is invariant under the exchange of $q_{\alpha,k}$ with $q_{\alpha,l}$. If we further choose χ_0 symmetric with respect to exchange of $q_{\alpha,k}$ and $q_{\alpha,l}$ we find that $\Psi'(t)$ is anti-symmetric with respect to $(\mathbf{z}\sigma)$. Thus it can be expanded in Slater-determinants of $(3+M)$-dimensional dressed orbitals $\varphi'(\mathbf{z}\sigma)$ (for the details see also Supplemental Material Sec. II). These observations make the application of well-established many-body methods such as the KS approach to TDDFT possible in a straightforward manner.

The KS approach to TDDFT [28] maps the interacting many-body problem of Eq. (3) to a non-interacting auxiliary problem, i.e., $\hat{H}'_{\text{KS}}(t) = \hat{T}' + \hat{V}'_{\text{KS}}(t)$. This auxiliary dressed KS system, usually given in terms of a Slater determinant $\Phi'(t)$ of dressed orbitals with spatial part $\varphi'_k(\mathbf{z},t)$, is enforced to generate the same (3+M)-dimensional expectation value

$$\begin{aligned}n'(\mathbf{z},t) &= \langle\Psi'(t)|\hat{n}'(\mathbf{z})|\Psi'(t)\rangle\\ &= N\sum_{\sigma_1,...,\sigma_N}\int \mathrm{d}^{(3+M)(N-1)}z|\Psi'(\mathbf{z}\sigma_1,...,\mathbf{z}_N\sigma_N)|^2,\end{aligned}$$

of the dressed density operator $\hat{n}'(\mathbf{z}) = \sum_{k=1}^{N}\delta(\mathbf{z}-\mathbf{z}_k)$. To ensure this, a mean-field exchange-correlation (Mxc) potential $v'_{\text{Mxc}}[\Psi'_0,\Phi'_0,n'] = v'[\Psi'_0,n'] - v'_{\text{s}}[\Phi'_0,n']$ is introduced, where $v'[\Psi'_0,n']$ and $v'_{\text{s}}[\Phi'_0,n']$ are the potentials that yield the density $n'(\mathbf{z},t)$ in an interacting respectively non-interacting dressed system starting from the initial states Ψ'_0 respectively Φ'_0. This leads to non-linear dressed single-particle equations

$$\mathrm{i}\tfrac{\partial}{\partial t}\varphi'_k(\mathbf{z},t) = \left[-\tfrac{1}{2}\nabla^2_{\mathbf{z}} + v'(\mathbf{z},t) + v'_{\text{Mxc}}[n'](\mathbf{z},t)\right]\varphi'_k(\mathbf{z},t),$$

where the exact dressed density is given by $n'(\mathbf{z},t) = \sum_{k=1}^{N}|\varphi'_k(\mathbf{z},t)|^2$. Similarly we can of course also set up a

dressed ground-state density-functional theory [29]. By construction the dressed density reduces to the exact electron density via $n(\mathbf{r},t) = \int \mathrm{d}^M q\, n'(\mathbf{z},t)$ and to the exact expectation value of $p_\alpha(t) = \int \mathrm{d}^{(3+M)} z\, \frac{q_\alpha}{\sqrt{N}} n'(\mathbf{z},t)$ (see Supplemental Material Sec. I).

In order for the dressed KS approach to be practical we need two things. The first is to be able to handle the dimensionality of the dressed orbitals. In most situations of cavity quantum electrodynamics only one mode is important, making the application of this approach only one dimension more expensive than the standard KS approach. On the other hand, if more than one cavity mode is important, we can adopt a reduction of the basis set similar to the calculations done in Ref. [3] for a multi-mode cavity, where only up to a few-photon states are considered. In the case that many photons are involved, a simple mean-field treatment within the standard KS approach becomes accurate again [16] and a dressed approach becomes less attractive. The other thing we need for the dressed KS approach to be practical is an approximation for the Mxc potential. In this regard it is interesting to compare the equations of motion for the physical and the dressed KS systems (for the derivations, and a more complete analysis, we refer to Supplemental Material Sec. III). The physical equation of motion for the density [13, 30], obtained by Heisenberg's equations, is given by

$$\begin{aligned}\tfrac{\partial^2}{\partial t^2} n(\mathbf{r},t) &= \nabla_{\mathbf{r}} \cdot [n(\mathbf{r},t) \nabla_{\mathbf{r}} v(\mathbf{r},t)] \\ &\quad - \nabla_{\mathbf{r}} \cdot [\mathbf{Q}(\mathbf{r},t) + \mathbf{F}_{\mathrm{dip}}(\mathbf{r},t) + \mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)],\end{aligned}$$

where $\mathbf{Q}(\mathbf{r},t) = \mathrm{i}\langle\Psi(t)|[\hat{T}+\hat{W}, \hat{J}(\mathbf{r})]|\Psi(t)\rangle$ is the physical momentum-stress and interaction-stress forces. In turn $\hat{J}(\mathbf{r}) = \frac{1}{2\mathrm{i}} \sum_{k=1}^N (\delta(\mathbf{r}-\mathbf{r}_k)\overrightarrow{\nabla}_{\mathbf{r}_k} - \overleftarrow{\nabla}_{\mathbf{r}_k}\delta(\mathbf{r}-\mathbf{r}_k))$ is the physical current operator. Further,

$$\begin{aligned}\mathbf{F}_{\mathrm{dip}}(\mathbf{r},t) &= -\sum_{\alpha=1}^M \boldsymbol{\lambda}_\alpha \langle\Psi(t)|(\boldsymbol{\lambda}_\alpha \cdot \sum_{k=1}^N \mathbf{r}_k)\hat{n}(\mathbf{r})|\Psi(t)\rangle \\ &= -\sum_{\alpha=1}^M \boldsymbol{\lambda}_\alpha \left(n(\mathbf{r}t)(\boldsymbol{\lambda}_\alpha \cdot \mathbf{r}) + 2\int \mathrm{d}\mathbf{r}' \rho_2(\mathbf{r},\mathbf{r}',t)(\boldsymbol{\lambda}_\alpha \cdot \mathbf{r}') \right), \\ \mathbf{F}_{\mathrm{lin}}(\mathbf{r},t) &= \sum_{\alpha=1}^M \boldsymbol{\lambda}_\alpha \langle\Psi(t)|\omega_\alpha p_\alpha \hat{n}(\mathbf{r})|\Psi(t)\rangle,\end{aligned}$$

are the forces the photons exert on the electron density [13]. Here $\rho_2(\mathbf{r},\mathbf{r}',t) = \frac{1}{2}\sum_{k\neq l}\langle\Psi(t)|\delta(\mathbf{r}-\mathbf{r}_k)\delta(\mathbf{r}'-\mathbf{r}_l)|\Psi(t)\rangle$ is the pair density. In contrast the dressed KS equation of motion (also for approximate v'_{Mxc}) reads,

$$\begin{aligned}\tfrac{\partial^2}{\partial t^2} n'(\mathbf{z},t) &= \nabla_{\mathbf{z}} \cdot [n'(\mathbf{z},t) \nabla_{\mathbf{z}} [v'(\mathbf{z},t) + v'_{\mathrm{Mxc}}(\mathbf{z},t)]] \\ &\quad - \nabla_{\mathbf{z}} \cdot \mathbf{Q}'_{\mathrm{KS}}(\mathbf{z},t), \end{aligned} \tag{4}$$

where $\mathbf{Q}'_{\mathrm{KS}}(\mathbf{z},t) = \mathrm{i}\langle\Phi'(t)|[\hat{T}', \hat{J}'(\mathbf{z})]|\Phi'(t)\rangle$ and $\hat{J}'(\mathbf{z}) = \frac{1}{2\mathrm{i}}\sum_{k=1}^N (\delta(\mathbf{z}-\mathbf{z}_k)\overrightarrow{\nabla}_{\mathbf{z}_k} - \overleftarrow{\nabla}_{\mathbf{z}_k}\delta(\mathbf{z}-\mathbf{z}_k))$. Inserting the expression for $v'(\mathbf{z},t)$ and integrating this equation over all q-coordinates, we find (again also for approximate v'_{Mxc})

$$\begin{aligned}\tfrac{\partial^2}{\partial t^2} n(\mathbf{r},t) &= \nabla_{\mathbf{r}} \cdot \left[n(\mathbf{r},t)\nabla_{\mathbf{r}} v(\mathbf{r},t) + \int \mathrm{d}^M q\, n'(\mathbf{z},t) \nabla_{\mathbf{r}} v'_{\mathrm{Mxc}}(\mathbf{z},t) \right] \\ &\quad - \nabla_{\mathbf{r}} \cdot \left[\mathbf{F}^{\mathrm{d}}_{\mathrm{dip,KS}}(\mathbf{r},t) + \mathbf{F}^{\mathrm{d}}_{\mathrm{lin,KS}}(\mathbf{r},t) + \mathbf{Q}^{\mathrm{d}}_{\mathrm{KS}}(\mathbf{r},t) \right], \end{aligned} \tag{5}$$

where $\mathbf{F}^{\mathrm{d}}_{\mathrm{dip,KS}}(\mathbf{r},t) = -\sum_{\alpha=1}^M \boldsymbol{\lambda}_\alpha n(\mathbf{r}t)(\boldsymbol{\lambda}_\alpha \cdot \mathbf{r})$, $\mathbf{F}^{\mathrm{d}}_{\mathrm{lin,KS}}(\mathbf{r},t) = \sum_{\alpha=1}^M \boldsymbol{\lambda}_\alpha \int \mathrm{d}^M q \frac{\omega_\alpha q_\alpha}{\sqrt{N}} n'(\mathbf{z},t) = \frac{1}{N}\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$ and $\mathbf{Q}^{\mathrm{d}}_{\mathrm{KS}}(\mathbf{r},t) = \mathrm{i}\langle\Phi'(t)|[\hat{T}, \hat{J}(\mathbf{r})]|\Phi'(t)\rangle$. That is, we get the $v(\mathbf{r},t)$ term as well as part of $\mathbf{F}_{\mathrm{dip}}(\mathbf{r},t)$ and $\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$ already from $v'(\mathbf{z},t)$, even if we set $v'_{\mathrm{Mxc}}(\mathbf{z},t) = 0$. We also get the kinetic forces $\mathbf{Q}^{\mathrm{d}}_{\mathrm{KS}}(\mathbf{r},t)$, and $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ then has to provide the rest of all the forces. To reproduce the exact $n'(\mathbf{z},t)$, $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ should of course reproduce the exact forces for $n'(\mathbf{z},t)$. However, more pragmatically, we often just want to get the right forces for $n(\mathbf{r},t)$ that different $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ can achieve, which may be simpler. To model the forces from $\hat{W}'$ that we are clearly missing, we can approximate them by their KS values [31] using

$$\nabla_{\mathbf{z}} \cdot [n'(\mathbf{z},t)\nabla_{\mathbf{z}} v'_{\mathrm{Mx}}(\mathbf{z},t)] = -\nabla_{\mathbf{z}} \cdot \mathrm{i}\langle\Phi'(t)|[\hat{W}', \hat{J}'(\mathbf{z})]|\Phi'(t)\rangle,$$

which we call mean-field exchange (Mx). In general we expect that designing $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ to approximate $\mathbf{F}_{\mathrm{dip}}(\mathbf{r},t)$ and $\mathbf{Q}(\mathbf{r},t)$ works the same as in the standard version of QEDFT KS, since they depend only on the electronic density. So the main difference is in how to approximate $\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$, and here we specifically get $\frac{1}{N}\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$ automatically. This allows us to scale the part of $v'(\mathbf{z},t)$ responsible for this term, $-\sum_{\alpha=1}^M \frac{\omega_\alpha q_\alpha}{\sqrt{N}}(\boldsymbol{\lambda}_\alpha \cdot \mathbf{r})$, by N to get the exact force expression for $\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$. This does, however, not automatically imply that we also get the exact forces, since only for the exact $n'(\mathbf{z},t)$ this would be true. Especially, if we only aim to get $n(\mathbf{r},t)$ right, the differences in $n'(\mathbf{z},t)$ must be compensated. Since $\mathbf{F}_{\mathrm{lin}}$ may start out zero and often depends more on the changes in $n'(\mathbf{z},t)$ (of a specific kind that contributes to the integral) than the initial $n'(\mathbf{z},t)$, the force can be very sensitive to this. Still, the fact that we can easily write the exact force expression in terms of $n'(\mathbf{z},t)$, which is hard in terms of the non-correlated quantities of standard QEDFT KS, indicates this may be a better starting point to approximate $\mathbf{F}_{\mathrm{lin}}$. Further note that the standard version of QEDFT KS usually translates to a special case of our dressed version (see Supplemental Material Section IV), allowing us to transfer (and extend) all standard approximations to here, but not vice versa.[1] A full analysis of all these aspects is beyond the scope of this work though. For the rest of this work we therefore stay within the Mx approximation.

[1] This is somewhat different though, as it reproduces the $n'(\mathbf{z},t)$ of $\Phi(t)\chi(t)$, instead of $\Psi(t)\chi(t)$, with $\Phi(t)$ the standard KS wave function. Nevertheless, both $n'(\mathbf{z},t)$ yield the same $n(\mathbf{r},t)$ and $p_\alpha(t)$.

In the following we consider two examples of two particles in a singlet state coupled to one mode with no external current $j(t) = 0$. Therefore we have $v'(\mathbf{r}, q, t) = v(\mathbf{r}, t) + \frac{1}{2}(\boldsymbol{\lambda}\cdot\mathbf{r})^2 + \frac{\omega^2}{2}q^2 - \frac{\omega}{\sqrt{2}}q(\boldsymbol{\lambda}\cdot\mathbf{r})$, and if we further take the spatial part of the dressed KS wave function to be described by a doubly-occupied dressed orbital $\varphi(\mathbf{r}, q, t)$, we have explicitly that

$$v'_{\mathrm{Mx}}(\mathbf{r}, q, t) = \int \mathrm{d}^3\mathbf{r}'\mathrm{d}q' \, |\varphi'(\mathbf{r}', q', t)|^2 \times \left[w(\mathbf{r}, \mathbf{r}') + (\boldsymbol{\lambda}\cdot\mathbf{r})(\boldsymbol{\lambda}\cdot\mathbf{r}') - \tfrac{\omega}{\sqrt{2}}q(\boldsymbol{\lambda}\cdot\mathbf{r}') - \tfrac{\omega}{\sqrt{2}}q'(\boldsymbol{\lambda}\cdot\mathbf{r})\right].$$

We note here, that if one employs this approximation for a $\varphi(\mathbf{r}, q, t)$ that is symmetric ($\varphi(\mathbf{r}, q, t) = \varphi(-\mathbf{r}, -q, t)$, like in our second example), only $w(\mathbf{r}, \mathbf{r}')$ contributes, and the coupling to the photon mode is only due to v'.

In our first example, we consider a simple two-site model with $v = w = 0$,

$$\hat{H} = -t\sum_{\sigma=1}^{2}(\hat{c}^\dagger_{1,\sigma}\hat{c}_{2,\sigma} + \hat{c}^\dagger_{2,\sigma}\hat{c}_{1,\sigma}) + \tfrac{1}{2}\left[-\tfrac{\partial^2}{\partial p^2} + \left(\omega p - \lambda\hat{d}\right)^2\right].$$

Here $\hat{c}^\dagger_{k,\sigma}$ and $\hat{c}_{k,\sigma}$ are fermionic creation and annihilation operators of the electrons at site k with spin σ, and $\hat{d} = \frac{1}{2}\sum_{\sigma=1}^{2}(\hat{c}^\dagger_{2,\sigma}\hat{c}_{2,\sigma} - \hat{c}^\dagger_{1,\sigma}\hat{c}_{1,\sigma})$ is the dipole operator. As initial state we choose some spin-singlet electronic state and the photon mode in its ground state, i.e., $\Psi_0 = \frac{1}{\sqrt{2}}\left(\sqrt{\frac{1}{4}}\,\hat{c}^\dagger_{1,\uparrow} + \sqrt{\frac{3}{4}}\,\hat{c}^\dagger_{2,\uparrow}\right)\left(\sqrt{\frac{1}{4}}\,\hat{c}^\dagger_{1,\downarrow} + \sqrt{\frac{3}{4}}\,\hat{c}^\dagger_{2,\downarrow}\right)|0\rangle_{\mathrm{e}}\otimes|0\rangle_{\mathrm{p}}$, where $|0\rangle_{\mathrm{e}}$ is the electronic vacuum and $|0\rangle_{\mathrm{p}}$ the photonic vacuum (the ground state of the harmonic oscillator in displacement representation). The resulting exact Rabi oscillation for $t = 0.5$, $\omega = 1$ and $\lambda = 0.01$ is depicted in Fig. 1 in red.

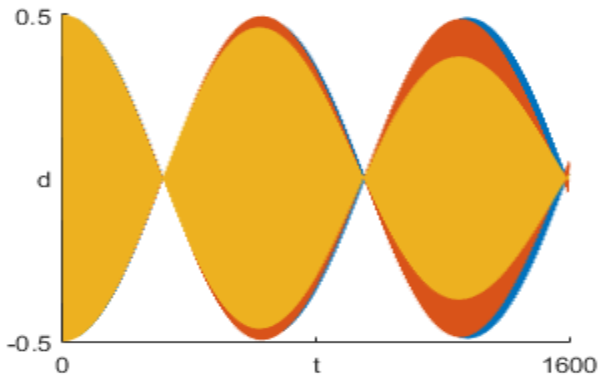

FIG. 1. (color online) The exact (red), TDOEP-(GW_0) approximated (orange) and dressed Mx approximated (blue) dipole moment of two electrons on two sites coupled to one mode.

Let us first compare to a relatively advanced functional within the standard KS scheme. We use a time-dependent optimized-effective potential (TDOEP) approach based on the exact-exchange energy expression of the Lamb shift [17], i.e., the second-order perturbation of the electronic system by the bare (W_0) photons. This functional accurately describes the correlated ground state [17, 18] but does not take into account the effect of novel light-matter correlated excited states (polaritons), i.e., the polaritonic eigenstates are not contributing to the non-adiabatic potential. Using the initial state Ψ_0 for the standard KS calculation, the mismatch in the higher eigenstates then leads to a beating in the (orange) dipole moment in Fig. 1, i.e., that represents the energetic mismatch between correlated and uncorrelated excited states. In contrast, if we employ the dressed Mx approximation (blue) with $\Phi'_0 = \Psi_0 \otimes |0\rangle_{p_2}$, where $|0\rangle_{p_2}$ is the ground state of $\hat{H}_{\mathrm{aux}}$, we qualitatively reproduce the right Rabi oscillations.

In our second example we go beyond a simple two-site model, and consider a one-dimensional model of helium using the soft Coulomb interaction, $v(x) = -2/\sqrt{x^2+1}$ and $w(x, x') = 1/\sqrt{|x-x'|^2+1}$ [32, 33], and with hard wall boundary conditions at $x = \pm 5$. Here we want to investigate photon-induced dynamics in real space. We first determine the ground-state of the "bare" helium (see Fig. 2 (a)), i.e, outside of the cavity, which for the exact calculation is a spin-singlet state ψ_0 and the excitation frequency for the lowest excited state is $\omega_1 = 0.58037$. Upon bringing helium into an empty cavity that is in resonance with the excitation energy $\omega = \omega_1$ with $\lambda = 0.1$, i.e., we use a tensor product $\Psi_0 = \psi_0 \otimes |0\rangle_{\mathrm{p}}$ and let it evolve with Eq. (1), we find the spontaneous emission behaviour in Fig. 2 (b).

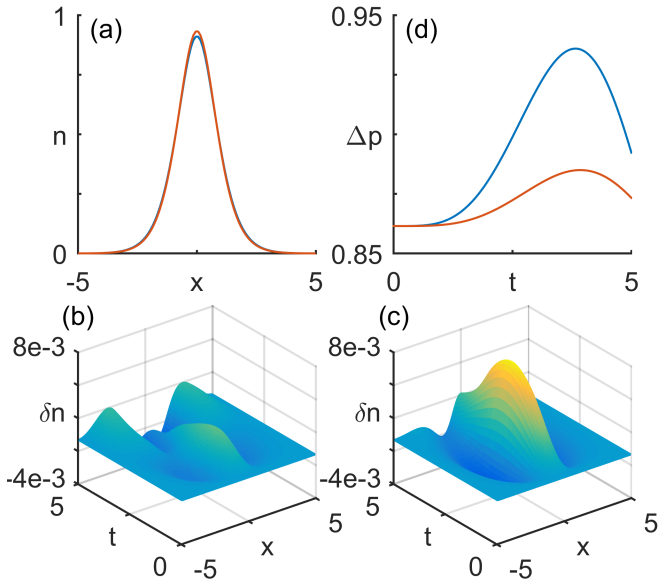

FIG. 2. (color online) (a) The exact (blue) and Mx (red) ground-state densities of the bare ($\lambda = 0$) one-dimensional Helium model. The corresponding changes in the (b) exact and (c) Mx density, $\delta n(x, t) = n(x, t) - n_0(x)$, when placed inside a cavity, and (d) the exact (blue) and Mx (red) field fluctuations.

However, since we only have one mode the emitted energy cannot dissipate but we will have a reoccurance time of the spontaneous emission [3]. Since the density is symmetric, the induced dipole moment is always zero and thus also the expectation value of the induced field. Hence, an approximation based on the photon-field expectation value $p(t)$ (as in the standard KS approach to coupled photon-matter systems) has a clear disadvantage in reproducing this spontaneous emission process, as also discussed in [3]. Now we investigate how the dressed KS scheme performs. First we solve for the bare KS ground state φ_0 using the Mx approximation for $\lambda = 0$ (see Fig. 2 (a)) and then set up the corresponding matter-photon initial state $\Phi_0' = \varphi_0 \otimes |0\rangle_{\rm p} \otimes |0\rangle_{\rm p_2}$. The photon field is then chosen with the same coupling strength λ and frequency ω as in the exact reference calculation above. In Fig. 2 (c) we can then see that a self-consistent propagation using the simple Mx approximation qualitatively recovers the photon-induced dynamics. Such a real-time and real-space test of an approximation is a much harder test than say, only looking at reduced quantities such as the energy or dipole moments [11]. Comparing to current studies about density-functional approximations [22, 34] the dressed Mx approximation performs indeed very good. If we drive the system, say with an external laser $v(\mathbf{r}, t)$ or an external current $j_\alpha(t)$, the results based on the dressed Mx approximation improve considerably as the external fields are taken into account exactly by the dressed KS approach.

Finally, let us look at the new types of observables that we can investigate. By construction both the exact and the dressed Mx approximation generate the exact $p(t) = 0$ in the above example. But we can also consider more complicated, approximate functionals. For instance, we can compare the field fluctuations $\Delta p(t) = \langle\Psi(t)|p^2|\Psi(t)\rangle - p^2(t)$ of the photon field to the approximated field-fluctuations $\int \mathrm{d}^2 z\, \frac{q^2}{2} n'(\mathbf{z}, t) - (\int \mathrm{d}^2 z\, \frac{q}{\sqrt{2}} n'(\mathbf{z}, t))^2$ from the dressed approach. Such observables become important, for instance, in the context of determining the photon statistics of an induced field [35]. Although the approximation to $\Delta p(t)$ is not supposed to become exact even for the *exact* dressed KS system, we find that we have again a qualitative agreement (see Fig. 2 (d)) in this otherwise very challenging observable [3].

To conclude, we have presented an alternative approach to coupled light-matter systems based on a higher-dimensional auxiliary system that explicitly correlates light and matter. We exemplified the novel possibilities by presenting an alternative KS construction. First calculations for real systems, as have been performed only recently in the context of the optimized effective potential approach [18], are currently on the way. It is important to note that any reasonably generic KS code can be used to perform the dressed KS calculations. Besides the extra dimensions, and slightly different $v'(\mathbf{z}, t)$ and $v'_{\rm Mxc}(\mathbf{z}, t)$, one has to select the proper orbitals (see Supplemental Material Sec. II). Also the observables are computed slightly differently. Finally, we point out that the dressed approach can equally-well be applied beyond the context of coupled light-matter systems. For instance, instead of treating photon modes strongly correlated with matter one could consider strong coupling to phonon modes. One can even think about modelling electron-electron correlation with the help of auxiliary degrees of freedom. This becomes specifically attractive for dispersion like interactions that are dipole-dipole interactions such as the van-der-Waals interaction and hence a dressed-state construction can follow the above ideas. These are future objectives that make this approach interesting also beyond the current physical context.

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Supplemental: Dressed-Orbital Approach to Cavity Quantum Electrodynamics and Beyond

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I. AUXILIARY COORDINATE TRANSFORMATION

Here we discuss the class of transformations allowing us to rewrite the physical problem in a higher-dimensional configuration space. We first specify this class abstractly, as many different transformations will do, as long as they satisfy a few basic properties. Although the specific choice of transformation does not matter, for completeness, and understanding, we also present one possible explicit option. Finally, we also show how the physical density $n(\mathbf{r},t)$ and photon-displacement $p_\alpha(t)$ are given in terms of the dressed density $n'(\mathbf{z},t)$.

A. Abstract Transformation

To establish a transformation from p_k to q_k coordinates that is orthogonal and also satisfies $p=\frac{1}{\sqrt{N}}(q_1+...+q_N)$, all we need to do is to require that the p-axis lies along the unit vector $\frac{1}{\sqrt{N}}(1,...,1)$ in q-space, while the other axes must simply be orthogonal to this and each other. If we further keep the same scale for all p_k and q_k axes, we have our desired transformation.

B. Explicit Transformation

To give one explicit special case of the above class of transformations, we may use the following for 4 electrons,

$$\begin{aligned} p &= \tfrac{1}{\sqrt{4}}\left(q_1+q_2+q_3+q_4\right),\\ p_2 &= \tfrac{1}{\sqrt{2}}\left(q_1-q_2\right),\\ p_3 &= \tfrac{1}{\sqrt{6}}\left(q_1+q_2-2q_3\right),\\ p_4 &= \tfrac{1}{\sqrt{12}}\left(q_1+q_2+q_3-3q_4\right), \end{aligned}$$

which clearly has an orthogonal transformation matrix. This transformation is easily generalised to any number of electrons using

$$p_k = \tfrac{1}{\sqrt{k^2-k}}\left(q_1+...+q_{k-1}-[k-1]q_k\right)$$

for $2\leq k\leq N$, of course, alongside $p=\frac{1}{\sqrt{N}}(q_1+...+q_N)$. The inverse of an orthogonal matrix is just the transpose, so for example for 4 electrons the inverse transformation (the one actually used to go from p- to q-coordinates) is

$$\begin{aligned} q_1 &= \tfrac{1}{\sqrt{4}}p+\tfrac{1}{\sqrt{2}}p_2+\tfrac{1}{\sqrt{6}}p_3+\tfrac{1}{\sqrt{12}}p_4\\ q_2 &= \tfrac{1}{\sqrt{4}}p-\tfrac{1}{\sqrt{2}}p_2+\tfrac{1}{\sqrt{6}}p_3+\tfrac{1}{\sqrt{12}}p_4\\ q_3 &= \tfrac{1}{\sqrt{4}}p-\tfrac{2}{\sqrt{6}}p_3+\tfrac{1}{\sqrt{12}}p_4\\ q_4 &= \tfrac{1}{\sqrt{4}}p-\tfrac{3}{\sqrt{12}}p_4 \end{aligned}$$

C. Wave Function Relations

For normalised $\Psi(\mathbf{r}_1\sigma_1,...,\mathbf{r}_N\sigma_N,p_1,...,p_M,t)$ and $\chi(p_{1,2},...,p_{M,N},t)$ and $\Psi'(\mathbf{z}_1\sigma_1,...,\mathbf{z}_N\sigma_N,t)\equiv\Psi\chi$ we find that

$$\begin{aligned} 1 &= \sum_{\sigma_1,...,\sigma_N}\int \mathrm{d}^{3N}r\,\mathrm{d}^{MN}p\,|\Psi\chi|^2\\ &= \sum_{\sigma_1,...,\sigma_N}\int \mathrm{d}^{(3+M)N}z\,|\Psi'|^2, \end{aligned}$$

so the total wave function Ψ' is also normalised. For Ψ' properly anti-symmetrized we further find that,

$$\begin{aligned} n(\mathbf{r},t) &= N\sum_{\sigma_1,...,\sigma_N}\int \mathrm{d}^{3(N-1)}r\,\mathrm{d}^{MN}p\,|\Psi\chi|^2\\ &= \int \mathrm{d}^M q\,\underbrace{N\sum_{\sigma_1,...,\sigma_N}\int \mathrm{d}^{(3+M)(N-1)}z\,|\Psi'|^2}_{=n'(\mathbf{z},t)},\\ p_\alpha(t) &= \sum_{\sigma_1,...,\sigma_N}\int \mathrm{d}^{3N}r\,\mathrm{d}^{MN}p\,p_\alpha|\Psi\chi|^2 \qquad (1)\\ &= \int \mathrm{d}^{3+M}z\,\tfrac{q_\alpha}{\sqrt{N}}n'(\mathbf{z},t). \end{aligned}$$

For later convenience, let us finally introduce the physical and dressed two-particle densities [1],

$$\begin{aligned} \rho_2(\mathbf{r},\mathbf{r}',t) &= \tfrac{N(N-1)}{2}\sum_{\sigma_1,...,\sigma_N}\int \mathrm{d}^{3(N-2)}r\mathrm{d}^M p\,|\Psi|^2,\\ \rho_2'(\mathbf{z},\mathbf{z}',t) &= \tfrac{N(N-1)}{2}\sum_{\sigma_1,...,\sigma_N}\int \mathrm{d}^{(3+M)(N-2)}z\,|\Psi'|^2, \end{aligned}$$

and note the relation $\rho_2(\mathbf{r},\mathbf{r}',t)=\int \mathrm{d}^M q\mathrm{d}^M q'\rho_2'(\mathbf{z},\mathbf{z}',t)$.

II. SYMMETRIES

The dressed Hamiltonian $\hat{H}'(t)$ is by design equivalent with the physical Hamiltonian when $v'(\mathbf{z},t)$, $w'(\mathbf{z},\mathbf{z}')$ and $\Psi'(t)$ correspond to a physical system. At the same time, it is also an extension to any $v'(\mathbf{z},t)$, $w'(\mathbf{z},\mathbf{z}')$ and $\Psi'(t)$.[1] The dressed KS formulation is based on this extension, and comprises a Φ'_0 and $v'_{\mathrm{KS}}(\mathbf{z},t)$ that recreate the $n'(\mathbf{z},t)$ of the physical $\Psi'(t)$, and thereby $n(\mathbf{r},t)$ as well as $p_\alpha(t)$. Unless all $\boldsymbol{\lambda}_\alpha = 0$, any dressed $\Phi'(t)$ therefore lacks certain fundamental symmetries that all $\Psi(t)\chi(t)$ possess. This difference of $\Phi'(t)$ and the $\Psi'(t)$ that it recreates the density of is compensated by $v'_{\mathrm{Mxc}}(\mathbf{z},t)$. It has to be build into approximate $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ to do the same. More generally we may consider any Φ'_0 and $v'_{\mathrm{KS}}(\mathbf{z},t)$ that recreate $n(\mathbf{r},t)$ and $p_\alpha(t)$, but different $n'(\mathbf{z},t)$.[2] A special case is the standard version of QEDFT [2] with KS wave function $\Phi(t)$, which corresponds to recreate the $n'(\mathbf{z},t)$ of $\Phi'(t) = \Phi(t)\chi(t)$. This $n'(\mathbf{z},t)$ separates in $n(\mathbf{r},t)\prod_{\alpha=1}^{M} n'_{q_\alpha}(q_\alpha,t)$. Therefore, while the dressed $v'_{\mathrm{KS}}(\mathbf{z},t)$ is generally correlated, that of standard KS separates in $v'_{\mathrm{KS},\mathbf{r}}(\mathbf{r},t) + \sum_{\alpha=1}^{M} v'_{\mathrm{KS},q_\alpha}(q_\alpha,t)$. Accordingly, the corresponding $\varphi'_k(\mathbf{z},t)$ orbitals of $\Phi'(t)$ usually also do not separate for the dressed KS approach, while the orbitals of the traditional KS approach do. Traditional KS has the advantage that $\Phi'(t)$ by design corresponds to a physical $\Phi(t)$, and so has the corresponding symmetries (and we can also solve it without the dressed description). However, if correlated orbitals capture the electron-photon correlation better, the corresponding $\Phi'(t)$ may well still be closer to $\Psi'(t)$ overall.

In this Section we discuss some of the symmetries that dressed Hamiltonians and wave functions corresponding to a physical system possess, and how approximations typically break these symmetries unless the Hamiltonian and initial state stay within the subspace corresponding to a physical system. We also show how the dressed KS Hamiltonian $\hat{H}'_{\mathrm{KS}}$ only shares all of these symmetries for $\boldsymbol{\lambda}_\alpha = 0$ (while that of standard KS always shares them), and how recovering the proper symmetries in this limit provides some insight into how to choose the KS initial state Φ'_0. Finally, we show that even if $\Phi'(t)$ lacks some symmetries of $\Psi'(t)$, many derived quantities like the corresponding electron n-body densities often recover these symmetries.

A. Symmetries of Dressed Physical Systems

Since electrons are fermions, $\hat{H}(t)$ is symmetric under exchange of $\mathbf{r}_k\sigma_k$ and $\mathbf{r}_l\sigma_l$. It is further symmetric under exchange of $q_{\alpha,k}$ and $q_{\alpha,l}$ when written in terms of $q_{\alpha,k}$ instead of the p_α, as the p_α by design have this symmetry. Since further all $\hat{H}_{\mathrm{aux},\alpha}$ share these exchange symmetries, $\hat{H}'(t) = \hat{H}(t) + \sum_{\alpha=1}^{M} \hat{H}_{\mathrm{aux},\alpha}$ inherits both these as well. This in turn also implies that $\hat{H}'(t)$ is symmetric under exchange of $\mathbf{z}_k\sigma_k$ and $\mathbf{z}_l\sigma_l$, since this is just a combination of the other symmetries.

Given some physical time-independent Hamiltonian $\hat{H}$, we may thus compute its $\mathbf{r}_k\sigma_k \leftrightarrow \mathbf{r}_l\sigma_l$ anti-symmetric eigenstates and the $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ symmetric eigenstates of the $\hat{H}_{\mathrm{aux},\alpha}$. These can then be combined into eigenstates of $\hat{H}'$ of the form $\Psi' = \Psi\prod_{\alpha=1}^{M}\chi_\alpha$ with both symmetries and therefore also with the dressed fermionic $\mathbf{z}_k\sigma_k \leftrightarrow \mathbf{z}_l\sigma_l$ anti-symmetry. Conversely, given a time-independent $\hat{H}'$ separating in a $\hat{H}$ and sum of $\hat{H}_{\mathrm{aux},\alpha}$ (using these depend on $\mathbf{r}_1\sigma_1, ..., \mathbf{r}_N\sigma_N, p_1, ..., p_M$ respectively $p_{\alpha,2}, ..., p_{\alpha,N}$), with proper exchange-symmetries, we may always pick all the relevant eigenstates to be of this particular form. An initial state Ψ'_0 separating into $\Psi_0\prod_{\alpha=1}^{M}\chi_{\alpha,0}$ and/or with (some of) the above symmetries, will thus also keep these properties through evolution under the exact $\hat{H}'(t)$. Note that the ground state of $\hat{H}_{\mathrm{aux},\alpha}$ is just a product of harmonic oscillator ground states $(\frac{\omega_\alpha}{\pi})^{1/4}\exp(-\frac{\omega_\alpha}{2}p_{\alpha,k}^2)$, since this simple product has the $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ symmetry. However, this is special to the harmonic oscillator ground states, and even for excited states it is more complicated.

Note that $\hat{V}'(t)$ and $\hat{W}'$ do not individually possess the $\mathbf{r}_k\sigma_k \leftrightarrow \mathbf{r}_l\sigma_l$ and $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ exchange symmetries, only the dressed $\mathbf{z}_k\sigma_k \leftrightarrow \mathbf{z}_l\sigma_l$ symmetry, in contrast to $\hat{V}(t)$ and $\hat{W}$. Further, they do not individually separate into two terms of $\mathbf{r}_1\sigma_1, ..., \mathbf{r}_N\sigma_N, p_1, ..., p_M$ and $p_{1,2}, ..., p_{M,N}$ like $\hat{H}'(t)$. These symmetries and the separation thus rely on a fine balance between $\hat{V}'(t)$ and $\hat{W}'$, which is rather unique to $v'(\mathbf{z},t)$ and $w'(\mathbf{z},\mathbf{z}')$ that correspond to a physical system. Approximations can therefore break these symmetries. The only case where $\hat{V}'(t)$ and $\hat{W}'$ have these extra properties is if all $\boldsymbol{\lambda}_\alpha = 0$.

For all $\boldsymbol{\lambda}_\alpha = 0$, $\hat{H}'$ further separates in electron and photon parts $\hat{H}' = \hat{H}'_{\mathrm{E}} + \sum_{\alpha=1}^{M}\hat{H}'_{\mathrm{P},\alpha}$, $\hat{H}'_{\mathrm{P},\alpha} = \hat{H}'_{p_\alpha} + \hat{H}_{\mathrm{aux},\alpha}$. Here $\hat{H}'_{p_\alpha} = -\frac{1}{2}\frac{\partial^2}{\partial p_\alpha^2} + \frac{1}{2}\omega_\alpha^2(p_\alpha - \bar{p}_\alpha)^2 - \frac{1}{2}\omega_\alpha^2\bar{p}_\alpha^2$ is a shifted harmonic oscillator shifted by $\bar{p}_\alpha = -\frac{j_\alpha}{\omega_\alpha^3}$. The separating eigenstates of $\hat{H}'_{\mathrm{P},\alpha}$ takes the form $\Phi'_{\mathrm{P},\alpha} = \varphi_{p_\alpha}(p_\alpha - \bar{p}_\alpha)\chi_\alpha$, where $\varphi_{p_\alpha}(p_\alpha)$ is a harmonic oscillator eigenstate, and χ_α still is an eigenstate of $\hat{H}_{\mathrm{aux},\alpha}$ with $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ symmetry (so both parts have this symmetry). In $q_{\alpha,k}$ coordinates, $\hat{H}'_{\mathrm{P},\alpha} = \sum_{k=1}^{N}\Big(-\frac{1}{2}\frac{\partial^2}{\partial q_{\alpha,k}^2} + \frac{1}{2}\omega_\alpha^2(q_{\alpha,k} - \frac{\bar{p}_\alpha}{\sqrt{N}})^2 - \frac{1}{2N}\omega_\alpha^2\bar{p}_\alpha^2\Big)$.

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[1] To characterise the subsets of $v'(\mathbf{z},t)$ and $w'(\mathbf{z},\mathbf{z}')$ corresponding to a physical system, note that these can be written in the form $v'(\mathbf{z},t) = v(\mathbf{r},t) + \sum_{\alpha=1}^{M}[\frac{1}{2}\omega_\alpha^2 q_\alpha^2 - \frac{\omega_\alpha}{\sqrt{N}}q_\alpha(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}) + \frac{1}{2}(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r})^2 + \frac{j_\alpha(t)q_\alpha}{\sqrt{N}\omega_\alpha}]$ and $w'(\mathbf{z},\mathbf{z}') = w(\mathbf{r},\mathbf{r}') + \sum_{\alpha=1}^{M}[-\frac{\omega_\alpha}{\sqrt{N}}q_\alpha(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}') - \frac{\omega_\alpha}{\sqrt{N}}q'_\alpha(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}) + (\boldsymbol{\lambda}_\alpha\cdot\mathbf{r})(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}')]$ for arbitrary $v(\mathbf{r},t)$, $w(\mathbf{r},\mathbf{r}')$, ω_α, $\boldsymbol{\lambda}_\alpha$ and $j_\alpha(t)$. The ω_α and $\boldsymbol{\lambda}_\alpha$ used must be identical for $v'(\mathbf{z},t)$ and $w'(\mathbf{z},\mathbf{z}')$ though, linking the two spaces (as also seen in Subsection A). The subset of Ψ'_0 corresponding to a physical Ψ_0 is characterised by separability, i.e., $\Psi'_0 = \Psi_0\chi_0$, where the Ψ_0 has $\mathbf{r}_k\sigma_k \leftrightarrow \mathbf{r}_l\sigma_l$ exchange anti-symmetry and the χ_0 has $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ symmetry.

[2] To be precise, there will be a difference in how well $\Phi'(t)$ directly reproduce other quantities not given in terms of $n(\mathbf{r},t)$ and $p_\alpha(t)$, but that is about the only difference.

Here the symmetric eigenstates are permanents Φ'_{q_α} of N orbitals, $\varphi'_{q_\alpha,k}(q_\alpha-\frac{\bar{p}_\alpha}{\sqrt{N}})$, where each orbital is a harmonic oscillator eigenstate that need not be the same for all k. Many of these permanents are degenerate, namely if the orbital excitation levels add to the same total. To find the eigenstates that further separate in $\varphi_{p_\alpha}(p_\alpha-\bar{p}_\alpha)\chi_\alpha$ (i.e., to find $\Phi'_{\mathrm{P},\alpha}$ in $q_{\alpha,k}$ coordinates), we need to form linear combinations of these degenerate Φ'_{q_α} permanents. It is beyond our scope to determine the coefficients of these linear combinations, since we are mainly interested in the ground state, or possibly low excitations of φ_{p_α}, where the coefficients are easy to find. The ground state is a product (a single permanent of identical orbitals) $\prod_{k=1}^N\varphi'_{q_\alpha}(q_{\alpha,k}-\frac{\bar{p}_\alpha}{\sqrt{N}})$ of harmonic oscillator ground states (since the ground state χ_α is a product of such). For low φ_{p_α} only a few coefficients need to be found.[3]

B. Symmetries of Kohn-Sham Systems

Since individually $\hat{V}'(t)$ and $\hat{W}'$ generally only possess $\mathbf{z}_k\sigma_k\leftrightarrow\mathbf{z}_l\sigma_l$ symmetry, dressed KS Hamiltonians $\hat{H}'_{\mathrm{KS}}(t)$ generally also only have this symmetry, while $\hat{H}'_{\mathrm{KS}}(t)$ that correspond to standard KS have all physical properties.[4] In Section IV, we will thus show that the standard KS Hamiltonian $\hat{H}'_{\mathrm{KS}}(t)$ separates in electron and photon parts, $\hat{H}'_{\mathrm{KS}}(t)=\hat{H}'_{\mathrm{KS,E}}(t)+\sum_{\alpha=1}^M\hat{H}'_{\mathrm{KS,P},\alpha}(t)$. The photon mode Hamiltonians $\hat{H}'_{\mathrm{KS,P},\alpha}(t)$ are as in the exact case, $\hat{H}'_{\mathrm{KS,P},\alpha}(t)=\hat{H}'_{\mathrm{P},\alpha}(t)$, only here $\bar{p}_\alpha(t)=-\frac{j_\alpha(t)}{\omega_\alpha^3}+\frac{\boldsymbol{\lambda}_\alpha\cdot\mathbf{R}(t)}{\omega_\alpha}$. They therefore have the same properties and eigenstates. The only case that the dressed $\hat{H}'_{\mathrm{KS}}(t)$ also have these properties is if all $\boldsymbol{\lambda}_\alpha=0$, where the dressed and standard KS descriptions usually coincide and both reduce to pure electronic KS as there is no electron-photon correlation.[5] Especially, $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ also reduces to the purely electronic $v'_{\mathrm{Hxc}}(\mathbf{r},t)$, as the only two-body part of $\hat{H}'(t)$ is purely electronic ($w'(\mathbf{z},\mathbf{z}')=w(\mathbf{r},\mathbf{r}')$). For $\boldsymbol{\lambda}_\alpha\neq0$ the dressed $\hat{H}'_{\mathrm{KS}}(t)$ does not separate at all. From the form of $\hat{H}'_{\mathrm{KS}}(t)$, we see that the $v'_{\mathrm{KS}}(\mathbf{z},t)$ of standard KS, or dressed with all $\boldsymbol{\lambda}_\alpha=0$, separates into $v'_{\mathrm{KS},\mathbf{r}}(\mathbf{r},t)+\sum_{\alpha=1}^M v'_{\mathrm{KS},q_\alpha}(q_\alpha,t)$. This is due to the fact that $n'(\mathbf{z},t)$ separates. In contrast, the dressed $n'(\mathbf{z},t)$ and therefore $v'_{\mathrm{KS}}(\mathbf{z},t)$ do not separate for $\boldsymbol{\lambda}_\alpha\neq0$ (so approximate $v'_{\mathrm{KS}}(\mathbf{z},t)$ should also only separate if all $\boldsymbol{\lambda}_\alpha=0$).[6]

Given some time-independent standard KS Hamiltonian $\hat{H}'_{\mathrm{KS}}$ (or dressed with all $\boldsymbol{\lambda}_\alpha=0$) we may thus compute the $\mathbf{r}_k\sigma_k\leftrightarrow\mathbf{r}_l\sigma_l$ anti-symmetric eigenstates Φ'_{E} of $\hat{H}'_{\mathrm{KS,E}}$. These take the form of a single (or a sum of a few) Slater determinant $\Phi'_{\mathbf{r}\sigma}$ of $\varphi'_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)$ orbitals since the KS electrons do not interact. We may again further compute the separating and $q_{\alpha,k}\leftrightarrow q_{\alpha,l}$ symmetric eigenstates $\Phi'_{\mathrm{P},\alpha}$ of $\hat{H}'_{\mathrm{KS,P},\alpha}$. We may then combine the Φ'_{E} and $\Phi'_{\mathrm{P},\alpha}$ to form the eigenstates $\Phi'=\Phi'_{\mathrm{E}}\prod_{\alpha=1}^M\Phi'_{\mathrm{P},\alpha}$ of $\hat{H}'_{\mathrm{KS}}$, which have all the exchange symmetries and separate in all the physical ways. In $\mathbf{z}\sigma$ coordinates the combined Φ' ground state is a single Slater determinant (or a sum of such if Φ'_{E} is a sum) of the combined orbitals $\varphi'_k(\mathbf{z}\sigma)=\varphi'_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)\prod_{\alpha=1}^M\varphi'_{q_\alpha}(q_\alpha-\frac{\bar{p}_\alpha}{\sqrt{N}})$. This follows as $\prod_{\alpha=1}^M\Phi'_{\mathrm{P},\alpha}=\prod_{\alpha=1}^M\prod_{k=1}^N\varphi'_{q_\alpha}(q_{\alpha,k}-\frac{\bar{p}_\alpha}{\sqrt{N}})$ in this case (as showed earlier), which is a common part in all terms of the Slater determinant for the photon part. In ground-state KS this is all we need. In general though, the eigenstates are linear combinations of M-fold sums $\sum_{\tau^1}\ldots\sum_{\tau^M}\Phi'_{\tau^1,\ldots,\tau^M}$ of Slater determinants $\Phi'_{\tau^1,\ldots,\tau^M}$. This follows since Φ'_{E} and each $\Phi'_{\mathrm{P},\alpha}$ is a linear combination of determinants $\Phi'_{\mathbf{r}\sigma}$ respectively permanents Φ'_{q_α}, and each determinant-permanents product $\Phi'_{\mathbf{r}\sigma}\prod_{\alpha=1}^M\Phi'_{q_\alpha}$ equals such an M-fold sum. The N orbitals of each of the Slater determinants $\Phi'_{\tau^1,\ldots,\tau^M}$ are given by $\varphi'_{\tau^1,\ldots,\tau^M,k}(\mathbf{z}\sigma)=\varphi'_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)\prod_{\alpha=1}^M\varphi'_{q_\alpha,\tau^\alpha_k}(q_\alpha-\frac{\bar{p}_\alpha}{\sqrt{N}})$. Each of the τ^α sums in the M-fold sum then runs over all permutations τ^α of the sequence $(1,\ldots,N)$, and acts

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[3] For example, for the second excited φ_p and ground state χ we get $\varphi_p\chi=\frac{1}{\sqrt{2}}[2\omega(p-\bar{p})^2-1](\frac{\omega}{\pi})^{N/4}\exp(-\frac{1}{2}\omega[(p-\bar{p})^2+\sum_{k=2}^N p_k^2])=\{\frac{1}{N}\sum_{k=1}^N\frac{1}{\sqrt{2}}[2\omega(q_k-\frac{\bar{p}}{\sqrt{N}})^2-1]+\frac{1}{\sqrt{2N}}\sum_{k\neq l=1}^N\sqrt{2\omega}(q_k-\frac{\bar{p}}{\sqrt{N}})\sqrt{2\omega}(q_l-\frac{\bar{p}}{\sqrt{N}})\}(\frac{\omega}{\pi})^{N/4}\exp(-\frac{1}{2}\omega\sum_{k=1}^N(q_k-\frac{\bar{p}}{\sqrt{N}})^2)$. This is a linear combination of the two Φ'_{q_α} permanents with excitations that add to 2; one with all orbitals in their ground state except one second excited orbital, and one with two first excited orbitals. In general, it is limited how many Φ'_{q_α} permanents are needed (i.e., how many combinations of excitations add to a fixed total).

[4] This comes back to the fact that the dressed $v'_{\mathrm{KS}}(\mathbf{z},t)=v'(\mathbf{z},t)+v'_{\mathrm{Mxc}}(\mathbf{z},t)$ is only of the form of Footnote 1 (with the common $\boldsymbol{\lambda}_\alpha=0$ as $w'(\mathbf{z},\mathbf{z}')=0$) for all $\boldsymbol{\lambda}_\alpha=0$, while that of standard KS always takes this specific form.

[5] For a separating eigenstate $\Psi'=\Psi'_{\mathrm{E}}\prod_{\alpha=1}^M\Phi'_{\mathrm{P},\alpha}$ the dressed and standard KS descriptions indeed coincide, as the corresponding standard KS $\Phi'=\Phi'_{\mathrm{E}}\prod_{\alpha=1}^M\Phi'_{\mathrm{P},\alpha}$ shares the same density $n'(\mathbf{z})$. For correlated Ψ'_0, the $n'(\mathbf{z},t)$, and hence the descriptions, differ though (and do not reduce to electronic KS), even if all $\boldsymbol{\lambda}_\alpha=0$.

[6] For the all $\boldsymbol{\lambda}_\alpha=0$ separating eigenstates $\Psi'=\Psi'_{\mathrm{E}}\prod_{\alpha=1}^M\Phi'_{\mathrm{P},\alpha}$, or the $\Phi'=\Phi'_{\mathrm{E}}\prod_{\alpha=1}^M\Phi'_{\mathrm{P},\alpha}$ of standard KS, $n'(\mathbf{z})$ clearly separates. In contrast it does not for the $\boldsymbol{\lambda}_\alpha\neq0$ correlated Ψ' eigenstates. The dressed Φ' that recreates a non-correlated $n'(\mathbf{z})$ therefore also has to be correlated (as a non-correlated Φ' would just give a non-correlated $n'(\mathbf{z})$). Now, since for all $\boldsymbol{\lambda}_\alpha=0$ the dressed and standard KS coincide, Φ' and $\varphi'(\mathbf{z}\sigma)$ separate in this case. Therefore, since the form of Φ' should not change for $\boldsymbol{\lambda}_\alpha\neq0$, only the $\varphi'(\mathbf{z}\sigma)$ change and the correlation of Φ' must come from the $\varphi'(\mathbf{z}\sigma)$. In turn this implies $v'_{\mathrm{KS}}(\mathbf{z},t)$ must get correlated. As a side remark, note that the separation of $v'_{\mathrm{KS}}(\mathbf{z},t)$ is directly related with the exchange symmetries. For example, for two electrons and one mode, $\hat{V}'_{\mathrm{KS}}(t)=v'_{\mathrm{KS}}(\mathbf{r}_1,q_1,t)+v'_{\mathrm{KS}}(\mathbf{r}_2,q_2,t)$. The only case in which $\hat{V}'_{\mathrm{KS}}(t)$ is symmetric under exchange of $\mathbf{r}_1\sigma_1$ and $\mathbf{r}_2\sigma_2$ or q_1 and q_2 is when $v'_{\mathrm{KS}}(\mathbf{r},q,t)$ separates into $v'_{\mathrm{KS},\mathbf{r}}(\mathbf{r},t)+v'_{\mathrm{KS,q}}(q,t)$. For $\hat{V}'_{\mathrm{KS}}(t)$ to separate in two terms of $\mathbf{r}_1\sigma_1,\mathbf{r}_2\sigma_2,p_1$ and $p_{1,2}$ further requires a suitable $v'_{\mathrm{KS,q}}(q,t)$, so the $v'(\mathbf{z},t)$ and $w'(\mathbf{z},\mathbf{z}')$ of Footnote 1 are indeed rather special to have these properties.

as a symmetrizer for the $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ exchange symmetry for the given α since it sums over all permutations of the $\varphi'_{q_\alpha,k}(q_\alpha - \frac{\bar{p}_\alpha}{\sqrt{N}})$ orbitals. In the special case that all orbitals in each permanent Φ'_{q_α} are the same, all the $\Phi'_{\tau^1,...,\tau^M}$ become identical, and the M-fold sum reduces to a single determinant of $\varphi'_k(\mathbf{z}\sigma) = \varphi'_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)\prod_{\alpha=1}^M \varphi'_{q_\alpha}(q_\alpha)$ orbitals similar to the ground-state case. Unlike the individual, degenerate $\Phi'_{\tau^1,...,\tau^M}$ (with only $\mathbf{z}_k\sigma_k \leftrightarrow \mathbf{r}_l\sigma_l$ anti-symmetry), the Φ' linear combinations of M-fold sums have all the exchange symmetries and separate in all the physical ways. They further yield a different expression for $n'(\mathbf{z})$ in terms of the orbitals. Therefore, although in principle one may also just use one of the $\Phi'_{\tau^1,...,\tau^M}$ for Φ' (if one only demands $\mathbf{z}_k\sigma_k \leftrightarrow \mathbf{z}_l\sigma_l$ anti-symmetry) we expect a simpler $v'_{\text{Mxc}}(\mathbf{z})$ using these Φ'. Note that if we use the full $v'_{\text{KS}}(\mathbf{z},t)$ to find the $\varphi'_k(\mathbf{z}\sigma)$ of a ground state Φ', as opposed to use that it separates and then determine $\varphi'_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)$ and $\varphi'_{q_\alpha}(q_\alpha)$ individually, we should still require the $\varphi'_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)$ part of all $\varphi'_k(\mathbf{z}\sigma)$ (of each $\Phi'_{\mathbf{z}\sigma}$) differ (since the orbitals of each $\Phi'_{\mathbf{r}\sigma}$ must still differ). The orbitals with lowest energy then all have the same ground state photon part and we get the $\varphi'_k(\mathbf{z}\sigma)$ of before. For excited state Φ' one has to identify all $\varphi'(\mathbf{z}\sigma)$ that correspond to a $\varphi'_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)\prod_{\alpha=1}^M\varphi'_{q_\alpha,\tau_k^\alpha}(q_\alpha)$ of the given Φ'.

To generalise this to non-zero $\boldsymbol{\lambda}_\alpha$ also for dressed $\hat{H}'_{\text{KS}}$, we may for the Φ' ground state still just use a single (or a simple sum of) determinant(s) of $\varphi'_k(\mathbf{z}\sigma)$ orbitals. Only, the orbitals do no longer separate and Φ' only has the $\mathbf{z}_k\sigma_k \leftrightarrow \mathbf{z}_l\sigma_l$ anti-symmetry. To recover the above form if we set all $\boldsymbol{\lambda}_\alpha = 0$, we should again select the orbitals with lowest energy that further have different electron parts, though as the orbitals no longer perfectly separate, we need to refine what exactly we mean by this. The optimal way to do this is beyond our scope, but it is usually easy as long as the interaction is not too strong. For excited Φ' one has to identify all $\varphi'(\mathbf{z}\sigma)$ that for $\boldsymbol{\lambda}_\alpha = 0$ correspond to a $\varphi'_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)\prod_{\alpha=1}^M\varphi'_{q_\alpha,\tau_k^\alpha}(q_\alpha)$ of Φ'. Using the Φ' linear combinations of M-fold sums with these orbitals we then recover all properties for $\boldsymbol{\lambda}_\alpha = 0$.

For propagation, $\Phi'(t)$ at least always stay $\mathbf{z}_k\sigma_k \leftrightarrow \mathbf{z}_l\sigma_l$ anti-symmetric and of a numerically efficient $\mathbf{z}\sigma$-orbital form. The initial state Φ'_0 may have further properties, but these are only kept under evolution using a standard $\hat{H}'_{\text{KS}}(t)$, or a dressed if all $\boldsymbol{\lambda}_\alpha = 0$.

Finally, note that even if $\Phi'(t)$ only has $\mathbf{z}_k\sigma_k \leftrightarrow \mathbf{z}_l\sigma_l$ anti-symmetry, many derived physical quantities such as all electron n-body densities and density matrices recover appropriate exchange symmetries due to this symmetry. For example, take a spin-independent expectation value $O(\mathbf{r}_1,...,\mathbf{r}_n,\{p_\beta\},t) = \langle\Phi'(t)|\hat{O}(\mathbf{r}_1,...,\mathbf{r}_n,\{p_\beta\})|\Phi'(t)\rangle$. In general it may depend on $0 \leq n \leq N$ spatial coordinates and a subset $\{p_\beta\}$ of the photon coordinates p_α. To evaluate it we may need a coordinate transformation. Since the RHS is typically a spin-summed integral of a $\mathbf{z}_k\sigma_k \leftrightarrow \mathbf{z}_l\sigma_l$ symmetric integrand, also the LHS has this symmetry. For the LHS this is equivalent with $\mathbf{r}_k \leftrightarrow \mathbf{r}_l$ symmetry, since all the p_β are symmetric under exchange of $q_{\beta,k}$ and $q_{\beta,l}$, so this part of the $\mathbf{z}_k\sigma_k \leftrightarrow \mathbf{z}_l\sigma_l$ exchange has no effect, and the two kinds of exchange are identical. Note the argument generalises trivially to spin-dependent operators. We only restricted us for notational simplicity. In general it is thus only internal quantities like $\rho'_2(\mathbf{z},\mathbf{z}',t)$ that lack the $\mathbf{r}_k \leftrightarrow \mathbf{r}_l$ (anti)-symmetry. Also, even if $\Phi'(t)$ does not separate in a $\Phi(t)\chi(t)$, the $O(\mathbf{r}_1,...,\mathbf{r}_n,\{p_\beta\},t)$ expectation values only depend on the coordinates of $\Phi(t)$.

III. EQUATIONS OF MOTION AND FORCES

In this Section we first present the equations of motion for $n(\mathbf{r},t)$ and $p_\alpha(t)$ in the physical coordinates, to obtain the physical forces we want to capture with the dressed KS scheme. We then present the equation of motion for $n'(\mathbf{z},t)$ in the auxiliary coordinates for general $v'(\mathbf{z},t)$ and $w'(\mathbf{z},\mathbf{z}')$, which has the very same structure as the usual divergence of local-forces equation. We then study the special case where $v'(\mathbf{z},t)$ and $w'(\mathbf{z},\mathbf{z}')$ correspond to a physical system characterised by a $v(\mathbf{r},t)$, $w(\mathbf{r},\mathbf{r}')$ and $j_\alpha(t)$, and again present the equations of motion for $n(\mathbf{r},t)$ and $p_\alpha(t)$. But this time in terms of the quantities of the dressed system. This then allows us to see how the dressed system produces the same forces as the physical system, and to establish *exact relations* between the dressed and physical forces. Finally, we again present the equations of motion for $n'(\mathbf{z},t)$, $n(\mathbf{r},t)$ and $p_\alpha(t)$, but this time in terms of the quantities of the dressed KS system, to see how the Mxc potential produces the different forces.

A. Physical Equations of Motion

We consider the physical electron-photon Hamiltonian

$$\hat{H}(t) = \sum_{k=1}^N \left[-\tfrac{1}{2}\nabla^2_{\mathbf{r}_k} + v(\mathbf{r}_k,t)\right] + \tfrac{1}{2}\sum_{k\neq l} w(\mathbf{r}_k,\mathbf{r}_l) \qquad (2)$$
$$+ \sum_{\alpha=1}^M\left[-\tfrac{1}{2}\tfrac{\partial^2}{\partial p_\alpha^2} + \tfrac{1}{2}\Big(\omega_\alpha p_\alpha - \boldsymbol{\lambda}_\alpha\cdot\sum_{k=1}^N\mathbf{r}_k\Big)^2 + \tfrac{j_\alpha(t)}{\omega_\alpha}p_\alpha\right].$$

The expectation values $n(\mathbf{r},t) = \langle\Psi(t)|\hat{n}(\mathbf{r})|\Psi(t)\rangle$ and $\mathbf{J}(\mathbf{r},t) = \langle\Psi(t)|\hat{J}(\mathbf{r})|\Psi(t)\rangle$ of the physical density and current operators $\hat{n}(\mathbf{r}) = \sum_{k=1}^N\delta(\mathbf{r}-\mathbf{r}_k)$ and $\hat{J}(\mathbf{r}) = \frac{1}{2\mathrm{i}}\sum_{k=1}^N(\delta(\mathbf{r}-\mathbf{r}_k)\overrightarrow{\nabla}_{\mathbf{r}_k} - \overleftarrow{\nabla}_{\mathbf{r}_k}\delta(\mathbf{r}-\mathbf{r}_k))$ then obey the continuity equation

$$\tfrac{\partial}{\partial t}n(\mathbf{r},t) = -\nabla_{\mathbf{r}}\cdot\mathbf{J}(\mathbf{r},t).$$

Taking the second time-derivative, we then arrive at the divergence of local-force equation [3]

$$\tfrac{\partial^2}{\partial t^2}n(\mathbf{r},t) = \nabla_{\mathbf{r}}\cdot[n(\mathbf{r},t)\nabla_{\mathbf{r}}v(\mathbf{r},t)]$$
$$-\nabla_{\mathbf{r}}\cdot[\mathbf{Q}_{\text{kin}}(\mathbf{r},t) + \mathbf{Q}_{\text{int}}(\mathbf{r},t) + \mathbf{F}_{\text{dip}}(\mathbf{r},t) + \mathbf{F}_{\text{lin}}(\mathbf{r},t)],$$

where $\mathbf{Q}_{\rm kin}(\mathbf{r},t) = \mathrm{i}\langle\Psi(t)|[\hat{T},\hat{J}(\mathbf{r})]|\Psi(t)\rangle$ and $\mathbf{Q}_{\rm int}(\mathbf{r},t) = \mathrm{i}\langle\Psi(t)|[\hat{W},\hat{J}(\mathbf{r})]|\Psi(t)\rangle = -2\int \mathrm{d}\mathbf{r}'\rho_2(\mathbf{r},\mathbf{r}',t)\nabla_{\mathbf{r}}w(\mathbf{r},\mathbf{r}')$ are the physical momentum-stress and interaction-stress forces, and,

$$\begin{aligned}\mathbf{F}_{\rm dip}(\mathbf{r},t) &= -\sum_{\alpha=1}^{M}\boldsymbol{\lambda}_\alpha\langle\Psi(t)|(\boldsymbol{\lambda}_\alpha\cdot\sum_{k=1}^{N}\mathbf{r}_k)\hat{n}(\mathbf{r})|\Psi(t)\rangle \\ &= -\sum_{\alpha=1}^{M}\boldsymbol{\lambda}_\alpha\left(n(\mathbf{r}t)(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}) + 2\int \mathrm{d}\mathbf{r}'\rho_2(\mathbf{r},\mathbf{r}',t)(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}')\right), \\ \mathbf{F}_{\rm lin}(\mathbf{r},t) &= \sum_{\alpha=1}^{M}\boldsymbol{\lambda}_\alpha\langle\Psi(t)|\omega_\alpha p_\alpha\hat{n}(\mathbf{r})|\Psi(t)\rangle,\end{aligned}$$

are the forces the photons exert on the electron density [2]. Here $\mathbf{F}_{\rm lin}(\mathbf{r},t)$ is due to the linear coupling between the displacement field and the electrons, while $\mathbf{F}_{\rm dip}(\mathbf{r},t)$ is due to the dipole self-interaction, and balances the linear coupling such that the resulting Hamiltonian stays bounded from below, i.e., allows for a ground state [4].

The physical displacement coordinates similarly satisfy the mode-resolved Maxwell equations [2]

$$\tfrac{\partial^2}{\partial t^2}p_\alpha(t) = -\omega_\alpha^2 p_\alpha(t) + \omega_\alpha\boldsymbol{\lambda}_\alpha\cdot\mathbf{R}(t) - \tfrac{j_\alpha(t)}{\omega_\alpha},$$

where $\mathbf{R}(t) = \int \mathrm{d}^3r\,\mathbf{r}n(\mathbf{r},t)$ is the total dipole.

B. Dressed Equations of Motion

We consider the dressed electron-photon Hamiltonian

$$\hat{H}'(t) = \sum_{k=1}^{N}\left[-\tfrac{1}{2}\nabla_{\mathbf{z}_k}^2 + v'(\mathbf{z}_k,t)\right] + \tfrac{1}{2}\sum_{k\neq l}w'(\mathbf{z}_k,\mathbf{z}_l). \quad (3)$$

The expectation values $n'(\mathbf{z},t) = \langle\Psi'(t)|\hat{n}'(\mathbf{z})|\Psi'(t)\rangle$ and $\mathbf{J}'(\mathbf{z},t) = \langle\Psi'(t)|\hat{J}'(\mathbf{z})|\Psi'(t)\rangle$ of the dressed density and current operators $\hat{n}'(\mathbf{z}) = \sum_{k=1}^{N}\delta(\mathbf{z}-\mathbf{z}_k)$ and $\hat{J}'(\mathbf{z}) = \frac{1}{2\mathrm{i}}\sum_{k=1}^{N}(\delta(\mathbf{z}-\mathbf{z}_k)\overrightarrow{\nabla}_{\mathbf{z}_k} - \overleftarrow{\nabla}_{\mathbf{z}_k}\delta(\mathbf{z}-\mathbf{z}_k))$ then obey the continuity equation

$$\tfrac{\partial}{\partial t}n'(\mathbf{z},t) = -\nabla_{\mathbf{z}}\cdot\mathbf{J}'(\mathbf{z},t).$$

Taking the second time-derivative, we then arrive at

$$\begin{aligned}\tfrac{\partial^2}{\partial t^2}n'(\mathbf{z},t) &= \nabla_{\mathbf{z}}\cdot[n'(\mathbf{z},t)\nabla_{\mathbf{z}}v'(\mathbf{z},t)] \\ &\quad - \nabla_{\mathbf{z}}\cdot[\mathbf{Q}'_{\rm kin}(\mathbf{z},t) + \mathbf{Q}'_{\rm int}(\mathbf{z},t)],\end{aligned} \quad (4)$$

where $\mathbf{Q}'_{\rm kin}(\mathbf{z},t) = \mathrm{i}\langle\Psi'(t)|[\hat{T}',\hat{J}'(\mathbf{z})]|\Psi'(t)\rangle$ and $\mathbf{Q}'_{\rm int}(\mathbf{z},t) = \mathrm{i}\langle\Psi'(t)|[\hat{W}',\hat{J}'(\mathbf{z})]|\Psi'(t)\rangle = -2\int \mathrm{d}\mathbf{z}'\rho'_2(\mathbf{z},\mathbf{z}',t)\nabla_{\mathbf{z}}w'(\mathbf{z},\mathbf{z}')$ are the dressed momentum-stress and interaction-stress forces [5].

C. Physical Dressed Equations of Motion

Substituting the expressions for $v'(\mathbf{z},t)$ and $w'(\mathbf{z},\mathbf{z}')$ in Eq. (4) for a physical system, we obtain the equation of motion for $n'(\mathbf{z},t)$ in terms of $v(\mathbf{r},t)$, $w(\mathbf{r},\mathbf{r}')$ and $j_\alpha(t)$. This special case applies only for $v'(\mathbf{z},t)$ and $w'(\mathbf{z},\mathbf{z}')$ that correspond to a physical system.

Integrating the resulting equation over all q_α coordinates, using that the integral of a divergence vanishes for a closed system, we again find the equation of motion for $n(\mathbf{r},t)$, but this time expressed in terms of the quantities of the extended system, i.e.,

$$\begin{aligned}\tfrac{\partial^2}{\partial t^2}n(\mathbf{r},t) &= \nabla_{\mathbf{r}}\cdot[n(\mathbf{r},t)\nabla_{\mathbf{r}}v(\mathbf{r},t)] \\ &\quad - \nabla_{\mathbf{r}}\cdot\left[\mathbf{Q}^{\rm d}_{\rm kin}(\mathbf{r},t) + \mathbf{Q}^{\rm d}_{\rm int}(\mathbf{r},t) + \mathbf{F}^{\rm d}_{\rm dip}(\mathbf{r},t) + \mathbf{F}^{\rm d}_{\rm lin}(\mathbf{r},t)\right].\end{aligned}$$

Here the dressed stress and photon-matter forces are

$$\begin{aligned}\mathbf{Q}^{\rm d}_{\rm kin}(\mathbf{r},t) &= \mathrm{i}\langle\Psi'(t)|[\hat{T},\hat{J}(\mathbf{r})]|\Psi'(t)\rangle \\ \mathbf{Q}^{\rm d}_{\rm int}(\mathbf{r},t) &= -2\int \mathrm{d}\mathbf{r}'\,\rho_2(\mathbf{r},\mathbf{r}',t)\nabla_{\mathbf{r}}w(\mathbf{r},\mathbf{r}') \\ &\quad + (N-1)\mathbf{F}^{\rm d}_{\rm lin}(\mathbf{r},t) \\ &\quad - 2\sum_{\alpha=1}^{M}\boldsymbol{\lambda}_\alpha\int \mathrm{d}\mathbf{r}'\rho_2(\mathbf{r},\mathbf{r}',t)(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}'), \\ \mathbf{F}^{\rm d}_{\rm dip}(\mathbf{r},t) &= -\sum_{\alpha=1}^{M}\boldsymbol{\lambda}_\alpha n(\mathbf{r},t)(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}), \\ \mathbf{F}^{\rm d}_{\rm lin}(\mathbf{r},t) &= \sum_{\alpha=1}^{M}\boldsymbol{\lambda}_\alpha\int \mathrm{d}^M q\tfrac{\omega_\alpha q_\alpha}{\sqrt{N}}n'(\mathbf{z},t),\end{aligned}$$

where we note the $2^{\rm nd}$ term $(N-1)\mathbf{F}^{\rm d}_{\rm lin}(\mathbf{r},t)$ of $\mathbf{Q}^{\rm d}_{\rm int}(\mathbf{r},t)$ relies on the $q_{\alpha,k}\leftrightarrow q_{\alpha,l}$ exchange symmetry to establish that $2\int \mathrm{d}\mathbf{r}'\mathrm{d}^M q\,\rho'_2(\mathbf{z},\mathbf{z}',t) = (N-1)n'(\mathbf{r},q'_1,...,q'_M,t)$. For later reference when we introduce approximations, we note this is the only property the above expressions rely on specific to the subset of physical $v'(\mathbf{r},t)$ and $w'(\mathbf{z},\mathbf{z}')$. Breaking this symmetry by, for example, adding a $\delta v'(\mathbf{z},t)$ without this symmetry, one has to replace the given term by $2\sum_{\alpha=1}^{M}\boldsymbol{\lambda}_\alpha\int \mathrm{d}^M q\mathrm{d}^{3+M}z'\,\frac{\omega_\alpha q'_\alpha}{\sqrt{N}}\rho'_2(\mathbf{z},\mathbf{z}',t)$, while all the other terms above remain unchanged.

Multiplying the equation of motion for $n'(\mathbf{z},t)$ by $\frac{q_\alpha}{\sqrt{N}}$, and integrating this over all $\mathbf{z}$ coordinates using Eq. (1), we again find the mode-resolved Maxwell equations,

$$\tfrac{\partial^2}{\partial t^2}p_\alpha(t) = -\omega_\alpha^2 p_\alpha(t) + \omega_\alpha\boldsymbol{\lambda}_\alpha\cdot\mathbf{R}(t) - \tfrac{j_\alpha(t)}{\omega_\alpha},$$

with a contribution from $v'(\mathbf{z},t)$ of

$$-\omega_\alpha^2 p_\alpha(t) + \tfrac{\omega_\alpha}{N}\boldsymbol{\lambda}_\alpha\cdot\mathbf{R}(t) - \tfrac{j_\alpha(t)}{\omega_\alpha},$$

and a contribution from $w'(\mathbf{z},\mathbf{z}')$ of

$$\tfrac{(N-1)\omega_\alpha}{N}\boldsymbol{\lambda}_\alpha\cdot\mathbf{R}(t).$$

In the physical coordinates the mode-resolved Maxwell equations instead originated from the second line of the physical Hamiltonian (2).

D. Comparison of Force Terms: Exact Relations

The equations of motion for $n(\mathbf{r},t)$ and $p_\alpha(t)$ in terms of the quantities of the physical and exact dressed system must of course be the same, as trivial to confirm for $p_\alpha(t)$. For the electron density $n(\mathbf{r},t)$ this implies that

$$\mathbf{Q}^{\mathrm{d}}_{\mathrm{kin}}(\mathbf{r},t)+\mathbf{Q}^{\mathrm{d}}_{\mathrm{int}}(\mathbf{r},t)+\mathbf{F}^{\mathrm{d}}_{\mathrm{dip}}(\mathbf{r},t)+\mathbf{F}^{\mathrm{d}}_{\mathrm{lin}}(\mathbf{r},t)= \mathbf{Q}_{\mathrm{kin}}(\mathbf{r},t)+\mathbf{Q}_{\mathrm{int}}(\mathbf{r},t)+\mathbf{F}_{\mathrm{dip}}(\mathbf{r},t)+\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t).$$

This we can also easily confirm by using that for a dressed system corresponding to a physical one, $\Psi'(t)=\Psi(t)\chi(t)$ (this also ensures equality of the physical and dressed descriptions of $n(\mathbf{r},t)$, $p_\alpha(t)$ and $\rho_2(\mathbf{r},\mathbf{r}')$), which establishes the following interesting relations,

$$\begin{aligned}
\mathbf{Q}^{\mathrm{d}}_{\mathrm{kin}}(\mathbf{r},t)&=\mathbf{Q}_{\mathrm{kin}}(\mathbf{r},t),\\
\mathbf{Q}^{\mathrm{d}}_{\mathrm{int}}(\mathbf{r},t)&=\mathbf{Q}_{\mathrm{int}}(\mathbf{r},t)+\tfrac{N-1}{N}\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)\\
&\quad-2\sum_{\alpha=1}^{M}\boldsymbol{\lambda}_\alpha\int\mathrm{d}\mathbf{r}'\,\rho_2(\mathbf{r},\mathbf{r}',t)(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}'),\\
\mathbf{F}^{\mathrm{d}}_{\mathrm{dip}}(\mathbf{r},t)&=\mathbf{F}_{\mathrm{dip}}(\mathbf{r},t)\\
&\quad+2\sum_{\alpha=1}^{M}\boldsymbol{\lambda}_\alpha\int\mathrm{d}\mathbf{r}'\,\rho_2(\mathbf{r},\mathbf{r}',t)(\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}'),\\
\mathbf{F}^{\mathrm{d}}_{\mathrm{lin}}(\mathbf{r},t)&=\tfrac{1}{N}\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t).
\end{aligned}$$

Here we used for $\mathbf{F}^{\mathrm{d}}_{\mathrm{lin}}(\mathbf{r},t)$ how p_α, $n(\mathbf{r},t)$ and $n'(\mathbf{z},t)$ are defined in Section I, as well as the exchange symmetries of physical $\Psi'(t)$. This provides insight into how the different forces get recast in the dressed system. For example, it shows that the kinetic terms are the same in both cases, and that $\mathbf{F}^{\mathrm{d}}_{\mathrm{lin}}(\mathbf{r},t)$ gives $\frac{1}{N}$ of $\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$, while $\mathbf{Q}^{\mathrm{d}}_{\mathrm{int}}(\mathbf{r},t)$ provides the rest. Further, $\mathbf{F}_{\mathrm{dip}}(\mathbf{r},t)$ comes from the $(\boldsymbol{\lambda}_\alpha\cdot\hat{\mathbf{R}})^2$ term in $\hat{H}(t)$, which can be split into a one-body and two-body part, i.e., a potential and interaction. The one-body part leads to $\mathbf{F}^{\mathrm{d}}_{\mathrm{dip}}(\mathbf{r},t)$ both in the physical and the dressed system, while the two-body part then gives $\mathbf{F}_{\mathrm{dip}}(\mathbf{r},t)-\mathbf{F}^{\mathrm{d}}_{\mathrm{dip}}(\mathbf{r},t)$, so the dipole self-interaction is indeed treated the same in the two formulations (also in the case of a KS calculation).

E. Dressed Kohn-Sham Equations of Motion

The equation of motion for the dressed KS density is,

$$\begin{aligned}
\tfrac{\partial^2}{\partial t^2}n'(\mathbf{z},t)&=\nabla_{\mathbf{z}}\cdot[n'(\mathbf{z},t)\nabla_{\mathbf{z}}[v'(\mathbf{z},t)+v'_{\mathrm{Mxc}}(\mathbf{z},t)]]\\
&\quad-\nabla_{\mathbf{z}}\cdot\mathbf{Q}'_{\mathrm{KS}}(\mathbf{z},t).
\end{aligned}$$

Here the Mxc potential v'_{Mxc} models the force differences between the interacting and non-interacting systems, to get the same forces in the two systems, and is given by

$$\begin{aligned}
\nabla_{\mathbf{z}}\cdot[n'(\mathbf{z},t)\nabla_{\mathbf{z}}v'_{\mathrm{Mxc}}(\mathbf{z},t)]&=\nabla_{\mathbf{z}}\cdot\big(\mathbf{Q}'_{\mathrm{kin,s}}(\mathbf{z},t)-\mathbf{Q}'_{\mathrm{kin}}(\mathbf{z},t)\big)\\
&\quad-\nabla_{\mathbf{z}}\cdot\mathbf{Q}'_{\mathrm{int}}(\mathbf{z},t).
\end{aligned}\tag{5}$$

Here $\mathbf{Q}'_{\mathrm{kin,s}}(\mathbf{z},t)=\mathrm{i}\langle\Phi'(t)|[\hat{T}',\hat{J}'(\mathbf{z})]|\Phi'(t)\rangle$.

Like in Section III C, we may again obtain the equations of motion for $n(\mathbf{r},t)$ and $p_\alpha(t)$ by integration (after multiplication by $\frac{q_\alpha}{\sqrt{N}}$ in the latter case). This time, however, expressed in terms of the quantities of the dressed KS system. Since the results in Section III C do not rely on specific properties of the subset of physical $v'(\mathbf{r},t)$ and $w'(\mathbf{z},\mathbf{z}')$, we may in fact even reuse the results from there for all but the $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ terms. The equations of motion that we get therefore also hold even for approximate $v'_{\mathrm{Mxc}}(\mathbf{z},t)$,

$$\begin{aligned}
\tfrac{\partial^2}{\partial t^2}n(\mathbf{r},t)&=\nabla_{\mathbf{r}}\cdot\Big[n(\mathbf{r},t)\nabla_{\mathbf{r}}v(\mathbf{r},t)+\int\mathrm{d}^M q\,n'(\mathbf{z},t)\nabla_{\mathbf{r}}v'_{\mathrm{Mxc}}(\mathbf{z},t)\Big]\\
&\quad-\nabla_{\mathbf{r}}\cdot\left[\mathbf{F}^{\mathrm{d}}_{\mathrm{dip,KS}}(\mathbf{r},t)+\mathbf{F}^{\mathrm{d}}_{\mathrm{lin,KS}}(\mathbf{r},t)+\mathbf{Q}^{\mathrm{d}}_{\mathrm{KS}}(\mathbf{r},t)\right],\\
\tfrac{\partial^2}{\partial t^2}p_\alpha(t)&=-\omega_\alpha^2p_\alpha(t)+\tfrac{\omega_\alpha}{N}\boldsymbol{\lambda}_\alpha\cdot\mathbf{R}(t)-\tfrac{j_\alpha(t)}{\omega_\alpha}\\
&\quad-\tfrac{1}{\sqrt{N}}\int\mathrm{d}\mathbf{z}\,n'(\mathbf{z},t)\partial_{q_\alpha}v'_{\mathrm{Mxc}}(\mathbf{z},t).
\end{aligned}$$

To reproduce the exact $n'(\mathbf{z},t)$ we of course need to design $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ to generate the right forces for $n'(\mathbf{z},t)$. However, more pragmatically we may also try to just get the right forces for $n(\mathbf{r},t)$ only, or for $n(\mathbf{r},t)$ and $p_\alpha(t)$, but we then have to compensate for the errors in $n'(\mathbf{z},t)$.

IV. RELATIONSHIP WITH STANDARD CAVITY QUANTUM ELECTRODYNAMICS KOHN-SHAM THEORY

The standard QEDFT KS Hamiltonian [2], with an explicit mean-field contribution, reads,

$$\begin{aligned}
\hat{H}_{\mathrm{KS}}(t)&=\sum_{k=1}^{N}\Big[-\tfrac{1}{2}\nabla^2_{\mathbf{r}_k}+v(\mathbf{r}_k,t)+v_{\mathrm{mxc}}(\mathbf{r}_k,t)\\
&\qquad+\sum_{\alpha=1}^{M}(\boldsymbol{\lambda}_\alpha\cdot\mathbf{R}(t)-\omega_\alpha p_\alpha(t))\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}_k\Big]\\
&\quad+\sum_{\alpha=1}^{M}\Big[-\tfrac{1}{2}\tfrac{\partial^2}{\partial p_\alpha^2}+\tfrac{1}{2}\omega_\alpha^2p_\alpha^2-\omega_\alpha p_\alpha\boldsymbol{\lambda}_\alpha\cdot\mathbf{R}(t)+\tfrac{j_\alpha(t)}{\omega_\alpha}p_\alpha\Big],
\end{aligned}$$

where we denote with $v_{\mathrm{mxc}}(\mathbf{r},t)$ the mean-field exchange-correlation potential of standard QEDFT KS theory with the mean-field coupling to the photon field made explicit. Further, $\mathbf{R}(t)$ and $p_\alpha(t)$ are the expectation values of $\hat{\mathbf{R}}$ and p_α. This leads to the single particle equations

$$\begin{aligned}
\mathrm{i}\partial_t\varphi_{\mathbf{r},k}(\mathbf{r},t)&=\Big[-\tfrac{1}{2}\nabla^2_{\mathbf{r}}+v(\mathbf{r},t)+v_{\mathrm{mxc}}(\mathbf{r},t)\\
&\qquad+\sum_{\alpha=1}^{M}(\boldsymbol{\lambda}_\alpha\cdot\mathbf{R}(t)-\omega_\alpha p_\alpha(t))\boldsymbol{\lambda}_\alpha\cdot\mathbf{r}\Big]\varphi_{\mathbf{r},k}(\mathbf{r},t),\\
\mathrm{i}\partial_t\varphi_{p_\alpha}(p_\alpha,t)&=\Big[-\tfrac{1}{2}\tfrac{\partial^2}{\partial p_\alpha^2}+\tfrac{1}{2}\omega_\alpha^2p_\alpha^2\\
&\qquad-\omega_\alpha p_\alpha\boldsymbol{\lambda}_\alpha\cdot\mathbf{R}(t)+\tfrac{j_\alpha(t)}{\omega_\alpha}p_\alpha\Big]\varphi_{p_\alpha}(p_\alpha,t),
\end{aligned}$$

and the force equations,

$$\frac{\partial^2}{\partial t^2} n(\mathbf{r},t) = \nabla_{\mathbf{r}} \cdot [n(\mathbf{r},t) \nabla_{\mathbf{r}} [v(\mathbf{r},t) + v_{\mathrm{mxc}}(\mathbf{r},t)] - \nabla_{\mathbf{r}} \cdot \left[n(\mathbf{r},t) \sum_{\alpha=1}^{M} \boldsymbol{\lambda}_\alpha (\omega_\alpha p_\alpha(t) - \boldsymbol{\lambda}_\alpha \cdot \mathbf{R}(t)) \right],$$

$$\frac{\partial^2}{\partial t^2} p_\alpha(t) = -\omega_\alpha^2 p_\alpha(t) + \omega_\alpha \boldsymbol{\lambda}_\alpha \cdot \mathbf{R}(t) - \frac{j_\alpha(t)}{\omega_\alpha},$$

where the prior includes the mean-field approximations to $\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$ and $\mathbf{F}_{\mathrm{dip}}(\mathbf{r},t)$, and the second is the exact mode-resolved Maxwell equations due to the mean-field. To solve these equations one may solve the mode-resolved Maxwell equations analytically, $p_\alpha(t) = p_{\alpha,0}\cos(\omega_\alpha t) + \frac{\dot{p}_{\alpha,0}}{\omega_\alpha}\sin(\omega_\alpha t) + \int_0^t \sin(\omega_\alpha(t-t'))(\boldsymbol{\lambda}_\alpha \cdot \mathbf{R}(t') - \frac{j_\alpha(t')}{\omega_\alpha^2})\mathrm{d}t'$, and substitute the results into the single particle equations for $\varphi_{\mathbf{r},k}(\mathbf{r},t)$.

This problem can also be translated into a dressed KS problem by exactly the same recipe as in the exact case. That is, we add $\hat{H}_{\mathrm{aux}}$ to $\hat{H}_{\mathrm{KS}}(t)$ and switch coordinates to $(q_{\alpha,1},...,q_{\alpha,N})$ for each mode to find $\hat{H}'_{\mathrm{KS}}(t) = \hat{T}' + \hat{V}'_{\mathrm{KS}}(t)$, with $v'_{\mathrm{KS}}(\mathbf{z},t) = v(\mathbf{r},t) + v_{\mathrm{mxc}}(\mathbf{r},t) + \sum_{\alpha=1}^{M}(\boldsymbol{\lambda}_\alpha \cdot \mathbf{R}(t) - \omega_\alpha p_\alpha(t))\boldsymbol{\lambda}_\alpha \cdot \mathbf{r} + \sum_{\alpha=1}^{M}\left[\frac{1}{2}\omega_\alpha^2 q_\alpha^2 - \frac{\omega_\alpha}{\sqrt{N}} q_\alpha \boldsymbol{\lambda}_\alpha \cdot \mathbf{R}(t) + \frac{j_\alpha(t)}{\sqrt{N}\omega_\alpha} q_\alpha\right]$. We get no $\hat{W}'_{\mathrm{KS}}$, so this is a non-interacting dressed KS Hamiltonian. Note that $v_{\mathrm{mxc}}(\mathbf{r},t)$ depends only on $\mathbf{r}$, as usual in standard KS, while the dressed $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ depends on the full $\mathbf{z}$.

We see that $\hat{H}'_{\mathrm{KS}}(t)$ indeed separates in electron and photon parts, $\hat{H}'_{\mathrm{KS}}(t) = \hat{H}'_{\mathrm{KS,E}}(t) + \sum_{\alpha=1}^{M} \hat{H}'_{\mathrm{KS,P},\alpha}(t)$, where $\bar{p}_\alpha(t) = -\frac{j_\alpha(t)}{\omega_\alpha^3} + \frac{\boldsymbol{\lambda}_\alpha \cdot \mathbf{R}(t)}{\omega_\alpha}$. The eigenstates then take the form discussed in Section II B.

In conclusion, standard cavity QEDFT KS is a special case of dressed KS, where one reproduces the $n'(\mathbf{z},t)$ of the standard KS system instead of the physical system (but the same $n(\mathbf{r},t)$ and $p_\alpha(t)$). It is best implemented the standard way for numerical efficiency. The explicitly correlated orbitals of the dressed scheme have a limited extra numerical cost, but may capture electron-photon correlation better.

V. OUTLOOK

In this supplemental material we have given many details of the properties of the dressed physical and KS systems. We have further highlighted how the dressed KS construction reproduces the exact density $n'(\mathbf{z},t)$ by emulating the missing forces of the dressed physical system and thereby that of the original physical system. Since the dressed KS system is based on explicitly correlated orbitals, approximations to the Mxc potential are automatically correlated (at the expense of violating certain physical symmetries). However, also the standard QEDFT KS construction can be recast in the dressed picture. This allows one to use standard functionals and investigate them in the dressed setting. This interesting option together with all the relations between the physical, dressed and KS forces is the subject of ongoing work to find more accurate functionals for dressed QEDFT as well as standard QEDFT.

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