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 many-body methods 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 approach. The resulting dressed Kohn-Sham scheme allows for straightforward non-adiabatic approximations to the unknown exchange-correlation potential that explicitly includes correlations. We illustrate this for simple model systems placed inside a high-Q optical cavity, and show also result for observables such as the photon-field fluctuations that are hard to capture in standard matter-photon Kohn-Sham. We finally highlight that the dressed-orbital approach extends 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, chemical reactions can be modified by strongly coupling molecules to the vacuum field of an optical cavity [5, 6], strong exciton-photon coupling in light-harvesting complexes is expected to modify energy transfer [7], the coupling to quantum matter can lead to attractive photons [8] and the ultra-strong coupling to artificial atoms in circuit quantum electrodynamics extends far beyond the perturbative regime [9]. 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) [10], an accurate and unbiased description of the physical situation calls for calculations of the coupled matter-photon systems from first principles [3, 4, 11, 12]. An appealing such first-principles method is an extension of densityfunctional theory to coupled matter-photon systems [13– 16, which is called quantum-electrodynamical densityfunctional theory. By reformulating the fully coupled fermion-boson problem in a formally exact quantumfluid 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 is 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 [17, 18]. First calculations for real molecules coupled to photons in and out of equilibrium [19–21] 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 [3, 9, 22, 23]. Alternative approximation schemes either rely on a different auxiliary system, such as the strictly-correlated electron system [24] or go beyond the single Slater-determinant Ansatz [25].

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 higherdimensional orbitals. Since this leads to a "fermionization" of the coupled fermion-boson system it allows us to employ fermionic many-body methods such as Greens' function techniques [26, 27] or density-functional methods. Here we exemplify the possibilities of this approach by applying it in the context of time-dependent densityfunctional theory (TDDFT) for the case of an electronic quantum system coupled to the photons of an optical cavity. For this dressed KS scheme already the simplest approximations are non-adiabatic and include explicit correlations that can otherwise only be captured by advanced functionals for the standard KS scheme.

We show how simple approximations in terms of dressed (mixed matter-photon) orbitals capture the right Rabi-oscillation induced by the photon-matter coupling for a Rabi model and the spontaneous emission of a "bare" model helium that is brought inside an optical cavity.

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, 12]). 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) [28, 29]. In this case the interacting Hamiltonian reads

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

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 λ_{α} coupled to the total dipole of the electronic system. Furthermore, the photon modes are allowed to couple to an external current $J_{\alpha}(t)$ [14, 29].

We first introduce auxiliary extra dimensions $(p_{\alpha,2},...,p_{\alpha,N})$ for each mode, to extend each p_{α} up to the electronic dimension N, 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 $\hat{H}_{\text{aux},\alpha} = \sum_{k=2}^{N} \left(-\frac{1}{2} \frac{\partial^2}{\partial p_{\alpha,k}^2} + \frac{\omega_{\alpha}^2}{2} p_{\alpha,k}^2\right)$. 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

$$p_{\alpha} = \frac{1}{\sqrt{N}} (q_{\alpha,1} + \dots + q_{\alpha,N}).$$
 (1)

This is the inverse of a center-of-mass coordinate transformation [30] (see Supplemental Material Sec. I for an explicit example). Introducing a (3 + M)-dimensional dressed vector of space and auxiliary photon coordinates $\mathbf{z} = (\mathbf{r}, q_1, ..., q_M)$, i.e., each fermion is now a quasiparticle (polariton), we can then rewrite $\hat{H}'(t)$ as

$$\hat{H}'(t) = \sum_{k=1}^{N} \left[-\frac{1}{2} \nabla_{\mathbf{z}_{k}}^{2} + v'(\mathbf{z}_{k}, t) \right] + \frac{1}{2} \sum_{k \neq l} w'(\mathbf{z}_{k}, \mathbf{z}_{l})$$
(2)
= $\hat{T}' + \hat{V}'(t) + \hat{W}'$.

Here
$$v'(\mathbf{z},t) = v(\mathbf{r},t) + \sum_{\alpha=1}^{M} \left[\frac{1}{2}\omega_{\alpha}^{2}q_{\alpha}^{2} - \frac{\omega_{\alpha}}{\sqrt{N}}q_{\alpha}(\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}) + \frac{1}{2}(\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r})^{2} + \frac{j_{\alpha}(t)q_{\alpha}}{\sqrt{N}\omega_{\alpha}}\right]$$
 and $w'(\mathbf{z}, \mathbf{z}') = w(\mathbf{r}, \mathbf{r}') + \sum_{\alpha=1}^{M} \left[-\frac{\omega_{\alpha}}{\sqrt{N}}q_{\alpha}(\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}')\right]$

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

$$\Psi'(\mathbf{z}_{1}\sigma_{1},...,\mathbf{z}_{N}\sigma_{N},t) =
\Psi(\mathbf{r}_{1}\sigma_{1},...,\mathbf{r}_{N}\sigma_{N},p_{1},...,p_{M},t)\chi(p_{1,2},...,p_{M,N},t),$$

is the normalized solution of the extended TDSE $\mathrm{i} \frac{\partial}{\partial t} \Psi'(t) = \hat{H}'(t) \Psi'(t)$ with $\chi(t)$ the state that evolves from an arbitrary, normalised initial state χ_0 under \hat{H}_{aux} . Since $\Psi(t)$ depends only on the coordinates defined in Eq. (1), it is invariant under exchange of $q_{\alpha,k}$ with $q_{\alpha,l}$. If we further pick χ_0 symmetric with respect to exchange of $\mathbf{q}_{\alpha,k}$ and $\mathbf{q}_{\alpha,l}$, $\Psi'(t)$ is anti-symmetric under exchange of $\mathbf{z}_k \sigma_k$ and $\mathbf{z}_l \sigma_l$. It can therefore be expanded in Slater-determinants of (3+M)-dimensional dressed orbitals $\varphi'(\mathbf{z}\sigma)$. These observations make the application of well-established many-body methods such as the KS approach to TDDFT possible. We refer to Supplemental Material Sec. II for further details. 1

The KS approach [31] maps the interacting many-body problem of Eq. (2) to a non-interacting auxiliary problem, i.e., $\hat{H}'_{\rm KS}(t) = \hat{T}' + \hat{V}'_{\rm KS}(t)$. 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

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

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'_{Mxc} is introduced that mimics the interactions. This leads to non-linear dressed single-particle equations

$$i\frac{\partial}{\partial t}\varphi_k'(\mathbf{z},t) = \left[-\frac{1}{2}\nabla_{\mathbf{z}}^2 + v'(\mathbf{z},t) + v'_{\text{Mxc}}(\mathbf{z},t)\right]\varphi_k'(\mathbf{z},t),$$

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 [32]. By construction the dressed density reduces to the exact electron density via $n(\mathbf{r},t) = \int d^M q \, n'(\mathbf{z},t)$ and to the exact expectation value of $p_{\alpha}(t) = \int d^{(3+M)}z \, \frac{q_{\alpha}}{\sqrt{N}} n'(\mathbf{z},t)$.

¹ Note that \hat{H}' also has many eigenstates not in the form $\Psi' = \Psi \chi$, which are not physical. To compute only the physical eigenstates directly from \hat{H}' , one therefore still has to enforce the separation in $\Psi \chi$, $\mathbf{r}_k \sigma_k \leftrightarrow \mathbf{r}_l \sigma_l$ anti-symmetry and $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ symmetry. Other methods must also be adjusted to respect these properties.

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 (i.e., strongly correlated with the matter), and the application of this approach is only one dimension more expensive than standard KS (i.e., [14]). This renders dressed KS especially appealing. On the other hand, if more than one cavity mode matters, 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 fewphoton states are considered. In the case that many photons are involved, a simple mean-field treatment within the standard KS approach becomes accurate again [17] and a dressed approach becomes less attractive. 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 [14, 33], obtained by Heisenberg's equations, is given by

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

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

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

are the forces the photons exert on the electron density [14]. 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,

$$\frac{\partial^2}{\partial t^2} n'(\mathbf{z}, t) = \nabla_{\mathbf{z}} \cdot \{ n'(\mathbf{z}, t) \nabla_{\mathbf{z}} [v'(\mathbf{z}, t) + v'_{\text{Mxc}}(\mathbf{z}, t)] \}$$
$$- \nabla_{\mathbf{z}} \cdot \mathbf{Q}'_{\text{KS}}(\mathbf{z}, t),$$

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

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

where $\mathbf{F}_{\mathrm{dip,KS}}^{\mathrm{d}}(\mathbf{r},t) = -\sum_{\alpha=1}^{M} \boldsymbol{\lambda}_{\alpha} n(\mathbf{r}t) (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}),$ $\mathbf{F}_{\mathrm{lin,KS}}^{\mathrm{d}}(\mathbf{r},t) = \sum_{\alpha=1}^{M} \boldsymbol{\lambda}_{\alpha} \int \mathrm{d}^{M} q \frac{\omega_{\alpha} q_{\alpha}}{\sqrt{N}} n'(\mathbf{z},t) = \frac{1}{N} \mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$ and $\mathbf{Q}_{\mathrm{KS}}^{\mathrm{d}}(\mathbf{r},t) = \mathrm{i} \langle \Phi'(t) | [\hat{T},\hat{\mathbf{j}}(\mathbf{r})] | \Phi'(t) \rangle$. That is, we get the $v(\mathbf{r},t)$ term as well as parts 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 $\mathbf{Q}_{\mathrm{KS}}^{\mathrm{d}}(\mathbf{r},t)$, 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 reproduce the exact forces for $n'(\mathbf{z},t)$. However, given our interest in physical observables, it is usually sufficient if we can get the right forces for $n(\mathbf{r},t)$, which different $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ can achieve. To model the forces from \hat{W}' that we are clearly missing, we can approximate them by their KS values [34] using

 $\nabla_{\mathbf{z}} \cdot [n'(\mathbf{z}, t) \nabla_{\mathbf{z}} v'_{\mathbf{M}_{\mathbf{z}}}(\mathbf{z}, t)] = -\nabla_{\mathbf{z}} \cdot i \langle \Phi'(t) | [\hat{W}', \hat{\mathbf{j}}'(\mathbf{z})] | \Phi'(t) \rangle,$ which we call mean-field exchange (Mx). In general, we expect that designing $v'_{\text{Mxc}}(\mathbf{z},t)$ to approximate $\mathbf{F}_{\text{dip}}(\mathbf{r},t)$ and $\mathbf{Q}(\mathbf{r},t)$ works the same as in standard KS, since they only depend on the electrons. So the main difference is in how to approximate $\mathbf{F}_{lin}(\mathbf{r},t)$, and here we specifically get $\frac{1}{N}\mathbf{F}_{\text{lin}}(\mathbf{r},t)$ from $v'(\mathbf{z},t)$. We thus need only approximate the remaining part. Also, to do this, we may try to scale the part of $v'(\mathbf{z},t)$ responsible for this $\frac{1}{N}$ contribution, $-\sum_{\alpha=1}^{M} \frac{\omega_{\alpha} q_{\alpha}}{\sqrt{N}} (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r})$, by N to get the exact force expression for $\mathbf{F}_{lin}(\mathbf{r},t)$. Keep in mind that this not necessarily leads to the exact forces though since this would demand the exact $n'(\mathbf{z},t)$, and this idea is only designed with $n(\mathbf{r},t)$ not $n'(\mathbf{z},t)$ in mind. Since $\mathbf{F}_{lin}(\mathbf{r},t)$ depends strongly on the electron-photon correlations in $n'(\mathbf{z},t)$ it can be very sensitive to this. We investigate this approximation further in Supplemental Material Sec. V, while we focus in the following on the Mx approximation. Also note that the standard version of KS translates to a special case of our dressed version (see Supplemental Material Sec. IV).² This allows us to rewrite all standard approximations as dressed, but not vice versa.

In the following we compare dressed Mx with available standard functionals for two examples both featuring two particles in a singlet state coupled to one mode with no external current J(t)=0. Therefore we have $v'(\mathbf{r},q,t)=v(\mathbf{r},t)+\frac{\omega^2}{2}q^2-\frac{\omega}{\sqrt{2}}q(\boldsymbol{\lambda}\cdot\mathbf{r})+\frac{1}{2}(\boldsymbol{\lambda}\cdot\mathbf{r})^2$, 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

$$\begin{aligned} v'_{\text{Mx}}(\mathbf{r}, q, t) &= \frac{1}{2} \int d^3 \mathbf{r}' \, n(\mathbf{r}', t) w(\mathbf{r}, \mathbf{r}') \\ &+ \frac{1}{2} \left\{ [\boldsymbol{\lambda} \cdot \mathbf{R}(t) - \omega p(t)] (\boldsymbol{\lambda} \cdot \mathbf{r}) - \frac{\omega}{\sqrt{2}} q \boldsymbol{\lambda} \cdot \mathbf{R}(t) \right\}. \end{aligned}$$

² This follows the exact same recipe adding \hat{H}_{aux} to $\hat{H}_{KS}(t)$ and using $\Phi'(t) = \Phi(t)\chi(t)$, with $\hat{H}_{KS}(t)$ and $\Phi(t)$ the standard KS Hamiltonian and wave function. It is somewhat different though, as it reproduces the $n'(\mathbf{z},t)$ of $\Phi'(t) = \Phi(t)\chi(t)$ instead of $\Psi'(t) = \Psi(t)\chi(t)$, but this $n'(\mathbf{z},t)$ also yields the same $n(\mathbf{r},t)$ and $p_{\alpha}(t)$.

Here $\mathbf{R}(t) = \int \mathrm{d}^3 r \, \mathbf{r} n(\mathbf{r},t)$ is the total dipole, and we recall $n(\mathbf{r},t)$ and p(t) are given in terms of $n'(\mathbf{z},t) = 2|\varphi'(\mathbf{z},t)|^2$. Interestingly, the second line equals half the standard KS mean-field contribution in this particular case (while $v'(\mathbf{z},t)$ gives a different approximation for the other half). The first line is the Hartree exact exchange potential $v_{\mathrm{Hx}}(\mathbf{r},t)$, which here equals half the Hartree potential. In symmetric cases (like our second example), also for more electrons and modes, $v'_{\mathrm{Mx}}(\mathbf{z},t) = v_{\mathrm{Hx}}(\mathbf{r},t)$, and the coupling to the photon mode is only due to $v'(\mathbf{z},t)$.

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

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

Here $\hat{c}_{k,\sigma}^{\dagger}$ and $\hat{c}_{k,\sigma}$ are fermionic creation and annihilation operators of the electrons at site k with spin σ , while $\hat{d} = \frac{1}{2} \sum_{\sigma=1}^2 (\hat{c}_{2,\sigma}^{\dagger} \hat{c}_{2,\sigma} - \hat{c}_{1,\sigma}^{\dagger} \hat{c}_{1,\sigma})$ is the dipole operator. As initial state we pick a spin-singlet electronic state and the photon mode in its uncoupled ground state, i.e., $\Psi_0 = \frac{1}{\sqrt{2}} \left(\sqrt{\frac{1}{4}} \, \hat{c}_{1,\uparrow}^{\dagger} + \sqrt{\frac{3}{4}} \, \hat{c}_{2,\uparrow}^{\dagger} \right) \left(\sqrt{\frac{1}{4}} \, \hat{c}_{1,\downarrow}^{\dagger} + \sqrt{\frac{3}{4}} \, \hat{c}_{2,\downarrow}^{\dagger} \right) |0\rangle_e \otimes |0\rangle_p$. Here $|0\rangle_e$ and $|0\rangle_p$ refer to the electronic and 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 orange.

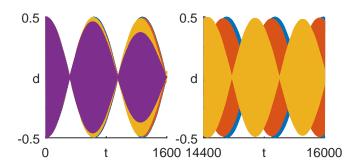


FIG. 1. (color online) The exact (orange), TDOEP- (GW_0) (lilac), standard Mx (blue) and dressed Mx (red) dipole moment of two electrons on two sites coupled to one mode.

We compare this to dressed Mx using $\Phi_0' = \Psi_0 \otimes |0\rangle_{p_2}$ as initial state, where $|0\rangle_{p_2}$ is the ground state of \hat{H}_{aux} , and to three cases within the standard KS scheme using Ψ_0 as initial state. Dressed Mx improves upon its direct counterpart standard Mx (blue) and standard mean-field (not plotted as it is visually indistinguishable from Mx for the given times), which especially manifests for later times. The third standard scheme is a time-dependent optimized-effective potential (TDOEP) GW_0 approach based on the exact-exchange energy expression of the Lamb shift [18]. This relatively advanced functional accurately describes the correlated ground state [18, 19], but does not self-consistently incorporate the polaritonic

eigenstates into the non-adiabatic potential, which manifests as a beating (lilac). In contrast the non-perturbative character of dressed Mx also allows the application to the ultra-strong coupling regime (e.g., circuit quantum electrodynamics), still with a consistent improvement over standard Mx and mean-field for all λ we tested.

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 $w(x, x') = 1/\sqrt{|x - x'|^2 + 1}$ [35, 36], and with hard wall boundary conditions at $x = \pm 5$. Here we want to investigate true quantum-induced light-matter dynamics in real space where classical mean-field methods will fail [37]. We first determine the exact ground-state ψ_0 of the "bare" helium (see Fig. 2 (a)), i.e, outside of the cavity, which is an electronic spin-singlet state. The excitation frequency for the lowest excited state is $\omega_1 = 0.58037$. Upon bringing helium into an empty cavity with $\omega = \omega_1$ and $\lambda = 0.1$, i.e., we use a tensor product $\Psi_0 = \psi_0 \otimes |0\rangle_p$, we find the spontaneous emission behaviour in Fig. 2 (b). As we only have one mode the emitted energy cannot dissipate and reoccurance takes place [3].

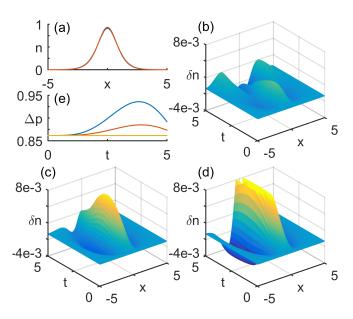


FIG. 2. (color online) (a) The exact (blue) and Mx (red) ground-state densities of the bare $(\lambda=0)$ one-dimensional Helium model. The corresponding changes in the (b) exact (c) dressed Mx and (d) standard Mx density, $\delta n(x,t) = n(x,t) - n_0(x)$, when placed inside a cavity, and (e) the exact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

To investigate how the dressed KS scheme performs we first 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 $\Phi_0'=\varphi_0\otimes|0\rangle_p\otimes|0\rangle_{p_2}$. 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 dressed Mx approximation qualitatively recovers the photon-induced dynamics and improves upon standard Mx in Fig. 2 (d). In contrast standard mean-field yields no density change as the electric field is always zero as the dipole moment is. 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 the dipole moment [12]. Note that within the standard KS scheme we solved the Coulomb interaction on an exact exchange level, and used $\Phi_0 = \varphi_0 \otimes |0\rangle_p$ as initial state. The difference between Mx and mean-field thus refers to the light-matter interaction.

By construction both the exact and the dressed Mx approximation generate the exact p(t)=0 in the given example. However, let us finally illustrate how we can also investigate observables such as the field fluctuations $\Delta p(t) = \langle \Psi(t)|p^2|\Psi(t)\rangle - p^2(t)$ of the photon field that are very challenging in standard KS [3]. The dressed approximation to the field fluctuations generally reads $\int \mathrm{d}^{3+M}z\,\frac{q_{\alpha}^2}{N}n'(\mathbf{z},t) + \int \mathrm{d}^{3+M}z\,\mathrm{d}^{3+M}z'\frac{2q_{\alpha}q'_{\alpha}}{N}\rho'_2(\mathbf{z}_1,\mathbf{z}_2,t) - p_{\alpha}^2(t)$, which in the given case reduces to $\int \mathrm{d}^2z\,\frac{q^2}{2}n'(\mathbf{z},t)$. We find again a qualitative agreement (see Fig. 2 (e)). In contrast standard Mx yields constant field fluctuations (as typical in standard KS as changes in the fluctuations originate from explicit light-matter correlation).

To conclude, we have presented a novel approach to coupled light-matter systems based on a higherdimensional auxiliary system that explicitly correlates light and matter, i.e., that considers polaritons as the underlying particles. We exemplified the possibilities by presenting an alternative KS construction and providing first benchmarks. We observed a consistent improvement of dressed Mx over its direct counterpart (standard Mx) as $\frac{1}{N}$ of $\mathbf{F}_{\text{lin}}(\mathbf{r},t)$ is covered directly by the known $v'(\mathbf{z},t)$ (for exact $n'(\mathbf{z},t)$) and only the remaining part is approximated. Scaling this $\frac{1}{N}$ fraction could lead to further improvements (see Supplemental Material Sec. V). Due to its non-perturbative character, our method remains applicable even for ultra-strong couplings, rendering this an ideal approach to low dimensional highly correlated problems such as circuit QED. Note that any reasonably generic KS code can be used to perform the dressed KS computations by working in higher dimensions **z** and by selecting the proper orbitals (see Supplemental Material Sec. II). 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 (polarons). One can even think about modelling electron-electron correlation with the help of auxiliary degrees of freedom, e.g., dispersion like interactions that are dipole-dipole interactions such as the van-der-Waals interaction. Such future objectives 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 are valid, as long as they satisfy a few basic properties. Although the specific choice of transformation does not matter we also present one possible explicit option for completeness and understanding. 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 $p=\frac{1}{\sqrt{N}}(q_1+\ldots+q_N)$, all we need to do is to require that the p-axis lies along the unit vector $\frac{1}{\sqrt{N}}(1,\ldots,1)$ 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,

$$p = \frac{1}{\sqrt{4}} (q_1 + q_2 + q_3 + q_4),$$

$$p_2 = \frac{1}{\sqrt{2}} (q_1 - q_2),$$

$$p_3 = \frac{1}{\sqrt{6}} (q_1 + q_2 - 2q_3),$$

$$p_4 = \frac{1}{\sqrt{12}} (q_1 + q_2 + q_3 - 3q_4),$$

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

$$p_k = \frac{1}{\sqrt{k^2 - k}} (q_1 + \dots + q_{k-1} - [k-1]q_k)$$

for $2 \le k \le N$, of course, alongside $p = \frac{1}{\sqrt{N}}(q_1 + \ldots + q_N)$. 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

$$q_{1} = \frac{1}{\sqrt{4}}p + \frac{1}{\sqrt{2}}p_{2} + \frac{1}{\sqrt{6}}p_{3} + \frac{1}{\sqrt{12}}p_{4}$$

$$q_{2} = \frac{1}{\sqrt{4}}p - \frac{1}{\sqrt{2}}p_{2} + \frac{1}{\sqrt{6}}p_{3} + \frac{1}{\sqrt{12}}p_{4}$$

$$q_{3} = \frac{1}{\sqrt{4}}p - \frac{2}{\sqrt{6}}p_{3} + \frac{1}{\sqrt{12}}p_{4}$$

$$q_{4} = \frac{1}{\sqrt{4}}p - \frac{3}{\sqrt{12}}p_{4}$$

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

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

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

$$n(\mathbf{r},t) = N \sum_{\sigma_1,\dots,\sigma_N} \int d^{3(N-1)} r \, d^{MN} p \, |\Psi\chi|^2$$

$$= \int d^M q \, N \sum_{\sigma_1,\dots,\sigma_N} \int d^{(3+M)(N-1)} z \, |\Psi'|^2,$$

$$= n'(\mathbf{z},t)$$

$$p_{\alpha}(t) = \sum_{\sigma_1,\dots,\sigma_N} \int d^{3N} r \, d^{MN} p \, p_{\alpha} |\Psi\chi|^2$$

$$= \int d^{3+M} z \, \frac{q_{\alpha}}{\sqrt{N}} n'(\mathbf{z},t).$$
(1)

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

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

which are related by $\rho_2(\mathbf{r}, \mathbf{r}', t) = \int d^M q d^M q' \rho'_2(\mathbf{z}, \mathbf{z}', t)$.

II. SYMMETRIES

In this Section we consider the space of all dressed Hamiltonians H'(t) characterised by $v'(\mathbf{z}, t)$ and $w'(\mathbf{z}, \mathbf{z}')$, which is displayed in Fig. 1.

```
All \hat{H}'[v'] with w'=0
All \hat{H}'[v', w']
 \begin{array}{c} \circ \mathbf{z}_k \sigma_k \leftrightarrow \mathbf{z}_l \sigma_l \text{ symmetry} \\ - \cup & - \cup & - \cup & - \cup & - \cup \\ - \cup & - \cup & - \cup & - \cup & - \cup \\ \end{array} ] - \text{Includes the dressed } \hat{H}'_{\text{KS}} \\ \text{Physical } \hat{H}'[v, w, \omega_{\alpha}, \boldsymbol{\lambda}_{\alpha}, \dot{J}_{\alpha}] \text{ physical } \hat{H}'[v, \omega_{\alpha}, \dot{J}_{\alpha}] \text{ with} 
 \rightarrow The physical properties: w' = 0, i.e., w = \lambda_{\alpha} = 0.

\begin{array}{c}
\text{The non-interacting} \\
\text{physical}
\end{array}

• \hat{H} + \sum_{\alpha} \hat{H}_{\text{aux},\alpha}
                                                                                                                         physical properties:
• \Psi' = \Psi \prod_{\alpha} \chi_{\alpha}
                                                                                                              • \hat{H}_{\rm E} + \sum_{\alpha} (\hat{H}_{p_{\alpha}} + \hat{H}_{\rm aux,\alpha})

• v'(\mathbf{z}) = v(\mathbf{r}) + \sum_{\alpha} v_{q_{\alpha}}(q_{\alpha})

• \Psi' = \Psi_{\rm E} \prod_{\alpha} \varphi_{p_{\alpha}} \chi_{\alpha}

• n'(\mathbf{z}) = n(\mathbf{r}) \prod_{\alpha} n_{q_{\alpha}}(q_{\alpha})
\circ \mathbf{r}_k \sigma_k \leftrightarrow \mathbf{r}_l \sigma_l symmetry
\circ q_{\alpha,k} \leftrightarrow q_{\alpha,l} symmetry
                                                                                                               – Includes the standard \hat{H}'_{KS}
```

FIG. 1. The space of all dressed Hamiltonians \hat{H}' . • Separation of \hat{H}' and the corresponding eigenstates Ψ' . \circ Exchange symmetries (silently also apply for all subsets \subset). Note that \hat{H} , \hat{H}_{E} , $\hat{H}_{p_{\alpha}}$, $\hat{H}_{aux,\alpha}$, Ψ , Ψ_{E} , $\varphi_{p_{\alpha}}$ and χ_{α} each satisfy all exchange symmetries (with $\mathbf{z}_k \sigma_k \leftrightarrow \mathbf{z}_l \sigma_l$ and $\mathbf{r}_k \sigma_k \leftrightarrow \mathbf{r}_l \sigma_l$ anti-symmetry for Ψ and $\Psi_{\rm E}$ but symmetry in all other cases).

Only a subset of these $\hat{H}'(t)$ corresponds to a physical Hamiltonian, $\hat{H}'(t) = \hat{H}(t) + \sum_{\alpha=1}^{M} \hat{H}_{\text{aux},\alpha}$, characterised by $v(\mathbf{r},t)$, $w(\mathbf{r},\mathbf{r}')$, ω_{α} , λ_{α} and $J_{\alpha}(t)$ through

$$v'(\mathbf{z},t) = v(\mathbf{r},t) + \sum_{\alpha=1}^{M} \left[\frac{1}{2} \omega_{\alpha}^{2} q_{\alpha}^{2} - \frac{\omega_{\alpha}}{\sqrt{N}} q_{\alpha} (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}) \right]$$

$$+ \frac{1}{2} (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r})^{2} + \frac{j_{\alpha}(t)q_{\alpha}}{\sqrt{N}\omega_{\alpha}} ,$$

$$w'(\mathbf{z}, \mathbf{z}') = w(\mathbf{r}, \mathbf{r}') + \sum_{\alpha=1}^{M} \left[-\frac{\omega_{\alpha}}{\sqrt{N}} q_{\alpha} (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}') \right]$$

$$- \frac{\omega_{\alpha}}{\sqrt{N}} q_{\alpha}' (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}) + (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}) (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}') ,$$

$$(3)$$

while all other pairs of $v'(\mathbf{z},t)$ and $w'(\mathbf{z},\mathbf{z}')$ correspond to a non-physical $\hat{H}'(t)$, and form an extension of the space. In Section II A we show that this subset of $\hat{H}'(t)$ exhibits certain properties that most non-physical $\hat{H}'(t)$ do not, and so we call these "the physical properties" in Fig. 1. That is, these properties rely on a fine balance between $v'(\mathbf{z},t)$ and $w'(\mathbf{z},\mathbf{z}')$ that approximations can break, e.g., it is important $v'(\mathbf{z},t)$ and $w'(\mathbf{z},\mathbf{z}')$ use the same ω_{α} and λ_{α} or $\hat{H}'(t)$ is not physical, and will break the properties. We further show that for a time-independent physical \hat{H}' the eigenfunctions separate in $\Psi' = \Psi \prod_{\alpha=1}^{M} \chi_{\alpha}$, where Ψ and each χ_{α} satisfy all the physical exchange symmetries. There are also many non-physical eigenstates not on this form, so to compute the physical eigenstates directly from H', one must enforce the physical properties, or compute all eigenstates and extract those on this form afterwards. The subset of Ψ'_0 corresponding to a physical Ψ_0 also satisfies the physical separability and exchange symmetries.

In this way, the dressed system is by design equivalent with the physical system when $v'(\mathbf{z},t)$, $w'(\mathbf{z},\mathbf{z}')$ and Ψ'_0 correspond to a physical system, but at the same time it is also an extension to any $v'(\mathbf{z},t)$, $w'(\mathbf{z},\mathbf{z}')$ and Ψ'_0 . To end Section II A we also discuss the all $\lambda_{\alpha} = 0$ case, where H'(t) separates in an electron and photon parts so KS should usually also reduce to purely electronic KS.

The dressed KS scheme is based on the extended space, and comprises a Φ'_0 and $v'_{KS}(\mathbf{z},t)$ that recreate the $n'(\mathbf{z},t)$ of the physical $\Psi'(t)$, and thereby $n(\mathbf{r},t)$ as well as $p_{\alpha}(t)$. In Section IIB we will thus show that any dressed $\Phi'(t)$ has none of the physical properties (unless all $\lambda_{\alpha} = 0$), and therefore does not correspond to any physical $\Phi'(t)$. This difference of $\Phi'(t)$ and the $\Psi'(t)$ that it recreates the density of is compensated by $v'_{\text{Mxc}}(\mathbf{z},t)$. It has to be build into approximate $v'_{\text{Mxc}}(\mathbf{z},t)$ to do the same though. More generally we may consider any Φ'_0 and $v'_{\text{KS}}(\mathbf{z},t)$ that recreate $n(\mathbf{r},t)$ and $p_{\alpha}(t)$, but different $n'(\mathbf{z},t)$. These are all equally interesting to the extent we can obtain them.¹ An important special case is the standard version [2] of cavity quantum-electrodynamical Kohn-Sham (QEKS). with KS wave function $\Phi(t)$. There is really no practical reason to perform standard KS in the dressed setting, yet it is worth our study to gain insight into dressed KS and for comparison. In Section IV we thus show that standard KS corresponds to recreate the $n'(\mathbf{z},t)$ of $\Phi'(t)$ $\Phi(t)\chi(t),$ and the standard KS Hamiltonian $\hat{H}'_{\mathrm{KS}}(t)$ takes the same form as a non-interacting $(w(\mathbf{r}, \mathbf{r}') = \lambda_{\alpha} = 0 =$ $w'(\mathbf{z}, \mathbf{z}')$) physical H'(t) defined by $v(\mathbf{r}, t)$, ω_{α} and $J_{\alpha}(t)$. That is, the standard KS potential can be written as that is, the standard HS possible $v'_{\text{KS}}(\mathbf{z},t) = v_{\text{KS},\mathbf{r}}(\mathbf{r},t) + \sum_{\alpha=1}^{M} \left[\frac{1}{2}\omega_{\alpha}^{2}q_{\alpha}^{2} + \frac{j_{\text{KS},\alpha}(t)q_{\alpha}}{\sqrt{N}\omega_{\alpha}}\right]$ to match the physical $v'(\mathbf{z},t) = v(\mathbf{r},t) + \sum_{\alpha=1}^{M} \left[\frac{1}{2}\omega_{\alpha}^{2}q_{\alpha}^{2} + \frac{j_{\alpha}(t)q_{\alpha}}{\sqrt{N}\omega_{\alpha}}\right]$. This form of the potentials ensures the further properties for non-interacting physical systems presented in Fig. 1, i.e., further separations in an electron and photon parts. That is, we see that $v'_{KS}(\mathbf{z},t)$ separates in an electron and photon parts and therefore so does $\hat{H}'_{\mathrm{KS}}(t)$ and so $\hat{H}_{\mathrm{KS}}(t)$. Thus $\hat{H}'_{KS}(t) = \hat{H}_{KS,E}(t) + \sum_{\alpha=1}^{M} [\hat{H}_{KS,p_{\alpha}}(t) + \hat{H}_{aux,\alpha}]$. Also, for a time-independent \hat{H}'_{KS} the eigenstates thus take the form $\Phi' = \Phi_E \prod_{\alpha=1}^{M} \varphi_{p_\alpha} \chi_\alpha$ for which the corresponding dressed densities separate $n'(\mathbf{z}) = n(\mathbf{r}) \prod_{\alpha=1}^{M} n_{q_{\alpha}}(q_{\alpha})^2$. This form tells us how to construct the Φ' eigenstates if we first compute the $\mathbf{r}\sigma$ -orbitals of $\Phi_{\rm E}$, the p_{α} -orbitals and the $p_{\alpha,k}$ -orbitals of χ_{α} using (the one-body form of) $\hat{H}_{KS,E}, \hat{H}_{KS,p_{\alpha}}$ respectively $\hat{H}_{aux,\alpha}$. The exception is that it is beyond our scope to determine the excited χ_{α} , which depend on $p_{\alpha,2},...,p_{\alpha,N}$ yet have $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ symmetry, as we are primarily interested in the ground state anyway. In Section II B we show that the Φ' can also be written as fixed linear combinations of determinants of $z\sigma$ -orbitals

¹ Essentially, the only minor difference is how well $\Phi'(t)$ directly

reproduce other quantities not given in terms of $n(\mathbf{r},t)$ and $p_{\alpha}(t)$. Note that $n_{q_{\alpha}}(q_{\alpha}) = \int \mathrm{d}^{3N}r\,\mathrm{d}^{NM-1}q|\Psi'|^2$ are not the photon mode densities $n_{p_{\alpha}}(p_{\alpha}) = \int \mathrm{d}^{3N}r\,\mathrm{d}^{M-1}p|\Psi|^2$.

(already in Section II A we study how $\varphi_{p_{\alpha}}\chi_{\alpha}$ look in $q_{\alpha,k}$ coordinates), which tells us how to construct the Φ' if we instead first compute the $\mathbf{z}\sigma$ -orbitals using the full H'_{KS} . We do not determine all the specific linear combinations, but we do show how it is straightforward in simple cases (like lowly excited $\Phi_{\rm E}$ and $\varphi_{p_{\alpha}}$) as are most relevant to us. In fact we only need the ground state in ground state KS, but low excitations of $\Phi_{\rm E}$ and $\varphi_{p_{\alpha}}$ may also be relevant to for example compute further properties, and are anyway worth a study to better understand the dressed structure. We show that the ground state is typically just a single determinant (or linear combination of a few differing only in their spin function). However, one has to use the right $\mathbf{z}\sigma$ -orbitals, which are often not those with lowest energy. For excited states the number of determinants grows fast with the excitation levels, but at least for few modes and low excitations the number of different orbitals remains manageable even so, allowing practical use in many cases. In dressed KS we may use the same linear combinations of determinants for the Φ' as we do in standard KS, only, as $v'_{KS}(\mathbf{z})$ gets correlated for $\lambda_{\alpha} \neq 0$ so do the $\mathbf{z}\sigma$ -orbitals. For example, we show that using the same form for the KS ground state in dressed and standard ground state KS ensures that both theories coincide and reduce to purely electronic KS for physical systems with all $\lambda_{\alpha} = 0$, as we show they should as they are to recreate the same $n'(\mathbf{z})$. If further $w(\mathbf{r}, \mathbf{r}') = 0$ for the physical system, this also ensures that the physical and both KS systems coincide. Using the same forms also for the excited states likewise ensures equivalence. These are strong arguments to use the standard KS forms for the Φ' also in dressed KS as other forms would break these equivalences and mimic the states by different forms instead of by the right ones. For those Φ' that consist of multiple determinants, the degeneracy of the determinants is often lifted in dressed KS though due to the correlated orbitals (an important exception is if the determinants only differ in their spin). These Φ' are therefore no longer truly eigenstates of H'_{KS} in dressed KS (unless all $\lambda_{\alpha} = 0$), so this is a price of we want the equivalences as clarified further in Section IIB. It is therefore not without issues to break the symmetries as dressed KS do, but dressed ground state KS seems to go free of these issues as long as Φ' is a single determinant (or the determinants only differ in their spin), as such Φ' are still eigenstates of H'_{KS} . Overall, standard KS thus has a clear advantage in that $\Phi'(t)$ by design is physical (and we can also solve it without the dressed description). However, if correlated orbitals, which rely on breaking the physicality,³ capture the electron-photon correlation better, the corresponding $\Phi'(t)$ may well still be closer to $\Psi'(t)$ overall. Also, lastly we show that even if $\Phi'(t)$ is not physical, many derived quantities like the corresponding electron n-body densities recover appropriate properties.

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. This establishes all the physical properties of physical $\hat{H}'(t)$ presented in Fig. 1.

Given some physical time-independent Hamiltonian 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 $H_{\text{aux},\alpha}$. 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. This verifies the form of the eigenstates stated in Fig. 1. Conversely, given a time-independent \hat{H}' separating in a \hat{H} and sum of $\hat{H}_{\text{aux},\alpha}$ (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 with (some of) the physical properties will thus also keep these under exact evolution by 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 excited χ_{α} are thus as said beyond our scope.

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 $\mathbf{z}_k \sigma_k \leftrightarrow \mathbf{z}_l \sigma_l$ symmetry, in contrast to $\hat{V}(t)$ and \hat{W} . Further, they do not individually separate into terms of $\mathbf{r}_1 \sigma_1, ..., \mathbf{r}_N \sigma_N, p_1, ..., p_M$ and $p_{\alpha,2}, ..., p_{\alpha,N}$ as $\hat{H}'(t)$ does. This is another example of these properties 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. The only case where $\hat{V}'(t)$ and \hat{W}' exhibit these physical properties individually is if all $\boldsymbol{\lambda}_{\alpha}=0$.

For all $\lambda_{\alpha} = 0$, \hat{H}' further separates in an electron and photon parts $\hat{H}' = \hat{H}_{\rm E} + \sum_{\alpha=1}^{M} \hat{H}'_{{\rm P},\alpha}$, $\hat{H}'_{{\rm P},\alpha} = \hat{H}_{p_{\alpha}} + \hat{H}_{{\rm aux},\alpha}$. 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

³ Insisting on a physical $v'_{\rm KS}({\bf z},t)$ in the sense of Eqs. (2) and (3) leads to standard KS (and separating orbitals), as $v'_{\rm KS}({\bf z},t)$ must (for a non-interacting $w'_{\rm KS}({\bf z},{\bf z}')=0$ KS system) then be of the non-interacting physical form of standard KS, for which only the standard KS $v_{\rm KS,r}({\bf r},t)$ and $\dot{J}_{\rm KS,\alpha}(t)$ recreate $n({\bf r},t)$ and $p_{\alpha}(t)$. The premise of using other $n'({\bf z},t)$ and correlated orbitals is thus to break the physical properties (see also Footnote 6, where we

argue that $v'_{\text{KS}}(\mathbf{z},t)$ must separate for $\hat{V}'_{\text{KS}}(t)$ to have the physical $\mathbf{r}_k \sigma_k \leftrightarrow \mathbf{r}_l \sigma_l$ and $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ exchange symmetries).

eigenstates of $\hat{H}'_{P,\alpha}$ take the form $\Phi'_{P,\alpha} = \varphi_{p_{\alpha}}(p_{\alpha} - \bar{p}_{\alpha})\chi_{\alpha}$, where $\varphi_{p_{\alpha}}(p_{\alpha})$ is a harmonic oscillator eigenstate, and χ_{α} still is an eigenstate of $H_{\mathrm{aux},\alpha}$ with $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ symmetry (so both parts have this symmetry). In $q_{\alpha,k}$ coordinates,
$$\begin{split} \hat{H}'_{\mathrm{P},\alpha} &= \sum_{k=1}^{N} \left[-\frac{1}{2} \frac{\partial^2}{\partial q_{\alpha,k}^2} + \frac{1}{2} \omega_{\alpha}^2 (q_{\alpha,k} - \frac{\bar{p}_{\alpha}}{\sqrt{N}})^2 - \frac{1}{2N} \omega_{\alpha}^2 \bar{p}_{\alpha}^2 \right]. \\ \text{Here the symmetric eigenstates are permanents } \Phi'_{q_{\alpha}} \text{ of } N \end{split}$$
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(and we normalise all the permanents in this work to 1). 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 $\Phi'_{P,\alpha}$ in $q_{\alpha,k}$ coordinates), we need to form linear combinations of these degenerate $\Phi'_{q_{\alpha}}$ permanents. It is beyond our scope to determine the coefficients of all these linear combinations, but it is easy to do for the low excitations we are mainly interested in. 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 then $\Phi'_{q_\alpha} = (\frac{\omega_\alpha}{\pi})^{N/4} \exp[-\frac{\omega_\alpha}{2} \sum_{k=1}^N (q_{\alpha,k} - \frac{\bar{p}_\alpha}{\sqrt{N}})^2]$ $= \left(\frac{\omega_{\alpha}}{\pi}\right)^{N/4} \exp\left\{-\frac{\omega_{\alpha}}{2} \left[(p_{\alpha} - \bar{p}_{\alpha})^2 + \sum_{k=2}^{N} p_{\alpha,k}^2 \right] \right\} = \varphi_{p_{\alpha}} \chi_{\alpha}.$ For low $\varphi_{p_{\alpha}}$ only a few coefficients need to be found.⁴

B. Symmetries of Kohn-Sham Systems

Recall from the introduction of this Section that the standard KS Hamiltonian $\hat{H}'_{\rm KS}(t)$ takes the same form as the physical Hamiltonian $\hat{H}'(t)$ for $w({\bf r},{\bf r}')={\pmb \lambda}_{\alpha}=0$, and therefore has all non-interacting physical properties. In Section IV we will thus show that $\hat{H}'_{\rm KS}(t)=\hat{H}_{\rm KS,E}(t)+\sum_{\alpha=1}^M\hat{H}'_{\rm KS,P,\alpha}(t)$, just as for such non-interacting $\hat{H}'(t)$. The photon mode Hamiltonians $\hat{H}'_{\rm KS,P,\alpha}(t)$ are the same as in the exact case for all ${\pmb \lambda}_{\alpha}=0$, $\hat{H}'_{\rm KS,P,\alpha}(t)=\hat{H}'_{\rm P,\alpha}(t)$, only here $\bar{p}_{\alpha}(t)=-\frac{j_{\alpha}(t)}{\omega_{\alpha}^3}+\frac{{\pmb \lambda}_{\alpha}\cdot{\bf R}(t)}{\omega_{\alpha}}$, where ${\bf R}(t)$ is the expectation value of the dipole operator $\hat{\bf R}=\sum_{k=1}^N{\bf r}_k$. They therefore have the same properties and eigenstates. The only case that the dressed $\hat{H}'_{\rm KS}(t)$ also has all these properties is if all ${\pmb \lambda}_{\alpha}=0$, in which case the dressed and standard KS descriptions usually coincide and reduce

to purely electronic KS as there is no electron-photon correlation.⁵ In this case $v'_{\text{Mxc}}(\mathbf{z},t)$ also reduces to the purely electronic Hartree exchange-correlation potential $v_{\rm Hxc}(\mathbf{r},t)$, as the two-body part of $\hat{H}'(t)$ becomes purely electronic $(w'(\mathbf{z}, \mathbf{z}') = w(\mathbf{r}, \mathbf{r}'))$. For $\lambda_{\alpha} \neq 0$, since $\hat{V}'(t)$ and W' individually only possess $\mathbf{z}_k \sigma_k \leftrightarrow \mathbf{z}_l \sigma_l$ symmetry, and the $n'(\mathbf{z},t)$ of $\Psi'(t)$ no longer separates, the dressed $\hat{H}'_{KS}(t)$ also only has this symmetry, and the $\hat{H}'_{KS}(t)$ and $v'_{KS}(\mathbf{z},t)$ do not separate.⁶ In dressed KS, approximate $v'_{\text{Mxc}}(\mathbf{z},t)$ should therefore also only separate if all $\lambda_{\alpha} = 0$. Given a time-independent standard KS Hamiltonian \hat{H}'_{KS} , or a dressed with all $\lambda_{\alpha} = 0$, we may thus compute the $\mathbf{r}_k \sigma_k \leftrightarrow \mathbf{r}_l \sigma_l$ anti-symmetric eigenstates Φ_E of $\hat{H}_{KS,E}$. These take the form of a single Slater determinant $\Phi_{\mathbf{r}\sigma}$ of $\varphi_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)$ orbitals (or a linear combination of a few such) as the KS electrons do not interact. We may again further compute the separating, $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ symmetric eigenstates $\Phi'_{P,\alpha}$ of the $H'_{KS,P,\alpha}$. 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}_{KS}' , which satisfy all the non-interacting physical properties. In $\mathbf{z}\sigma$ coordinates the combined Φ' ground state is a single Slater determinant of the combined orbitals $\varphi'_k(\mathbf{z}\sigma) =$ $\varphi_{{f r}\sigma,k}({f r}\sigma)\prod_{lpha=1}^M \varphi_{q_lpha}'(q_lpha-{ar p_lpha\over\sqrt N})$ (or a linear combination of such determinants in case $\Phi_{\rm E}$ is a linear combination).

⁴ For example, for the second excited φ_p and ground state χ we get $\varphi_p \chi = \frac{1}{\sqrt{2}} [2\omega(p-\bar{p})^2-1] (\frac{\omega}{\pi})^{N/4} \exp\{-\frac{1}{2}\omega[(p-\bar{p})^2+\sum_{k=2}^N p_k^2]\} = \{\frac{1}{N}\sum_{k=1}^N \frac{1}{\sqrt{2}} [2\omega(q_k-\frac{\bar{p}}{\sqrt{N}})^2-1] + \frac{1}{\sqrt{2}N}\sum_{k\neq l=1}^N \sqrt{2\omega}(q_k-\frac{\bar{p}}{\sqrt{N}}) (\frac{\omega}{\sqrt{N}})^{N/4} \exp[-\frac{1}{2}\omega\sum_{k=1}^N (q_k-\frac{\bar{p}}{\sqrt{N}})^2].$ This is a linear combination of the two Φ'_{qα} 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, the number of Φ'_{qα} permanents is given by the integer partition function p(Q) of the total excitation level Q (or is less for low N). This is easily managed for low excitations, and while p(Q) grows as $\exp(\pi\sqrt{\frac{2}{3}Q})$ asymptotically the number of different $\varphi'_{q\alpha,k}$ orbitals remains only Q+1, allowing further practical uses.

⁵ For a separating eigenstate $\Psi' = \Psi_E \prod_{\alpha=1}^{M} \Phi'_{P,\alpha}$ the dressed and standard KS descriptions indeed coincide, as the corresponding standard KS $\Phi' = \Phi_E \prod_{\alpha=1}^{M} \Phi'_{P,\alpha}$ shares the same density $n'(\mathbf{z})$. As standard KS reduces to purely electronic, so does dressed KS. 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 $\lambda_{\alpha} = 0$. ⁶ It would be surprising if $v'_{KS}(\mathbf{z},t) = v'(\mathbf{z},t) + v'_{Mxc}(\mathbf{z},t)$ would separate or be on the form of Eq. (2) (with the common $\lambda_{\alpha} = 0$ of Eqs. (2) and (3) as $w'_{KS}(\mathbf{z}, \mathbf{z}') = 0$) as $v'(\mathbf{z}, t)$ only separates and is on this form for all $\lambda_{\alpha} = 0$. Likewise, it would be surprising if $\hat{V}'_{\mathrm{KS}}(t) = \hat{V}'(t) + \hat{V}'_{\mathrm{Mxc}}(t)$ had further properties than $\hat{V}'(t)$ has. To argue more strictly for most of these facts, we first recall that for the all $\lambda_{\alpha} = 0$ separating eigenstates $\Psi' = \Psi_{\rm E} \prod_{\alpha=1}^{M} \Phi'_{\rm P,\alpha}$, or the $\Phi' = \Phi_{\rm E} \prod_{\alpha=1}^{M} \Phi'_{{\rm P},\alpha}$ of standard KS, $n'({\bf z})$ clearly separates. In contrast it does not for the $\lambda_{\alpha} \neq 0$ correlated Ψ' eigenstates. The dressed Φ' that recreates a correlated $n'(\mathbf{z})$ therefore also has to be correlated (since a non-correlated Φ' would just give a non-correlated $n'(\mathbf{z})$). Now, since for all $\lambda_{\alpha} = 0$ we want dressed and standard KS to coincide, Φ' and $\varphi'(\mathbf{z}\sigma)$ separate in this case. Therefore, since the form of Φ' should not change for $\lambda_{\alpha} \neq 0$, only the $\varphi'(\mathbf{z}\sigma)$, the correlation of Φ' must come from the $\varphi'(\mathbf{z}\sigma)$ get correlated. In turn this implies $v'_{\rm KS}({\bf z},t)$ must get correlated. Note that the separation or correlation of $v_{\rm KS}'(\mathbf{z},t)$ is also directly related with the exchange symmetries. For example, for two electrons and one mode, $\hat{V}_{\rm KS}'(t) = v_{\rm KS}'(\mathbf{r}_1,q_1,t) + v_{\rm KS}'(\mathbf{r}_2,q_2,t)$. 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 $v_{\rm KS,r}({f r},t)+v_{\rm KS,q}'(q,t).$ This also implies that no dressed scheme can have correlated orbitals, yet keep the exchange symmetries, as used in Footnote 3. For the $\hat{V}'_{KS}(t)$ in the example 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 $v'_{\mathrm{KS},\mathbf{q}}(q,t).$ Although we do not show it, it is thus highly unlikely that $\hat{V}'_{KS}(t)$ separates like this. The physical $v'(\mathbf{z}, t)$ and $w'(\mathbf{z}, \mathbf{z}')$ of Eqs. (2) and (3) are thus very special to have the discussed physical properties.

This follows as $\prod_{\alpha=1}^M \Phi'_{P,\alpha} = \prod_{\alpha=1}^M \prod_{k=1}^N \varphi'_{q_\alpha} (q_{\alpha,k} - \frac{\bar{p}_\alpha}{\sqrt{N}})$ 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 $C \sum_{\tau^1} \dots \sum_{\tau^M} \Phi'_{\tau^1,\dots,\tau^M}$ of Slater determinants $\Phi'_{\tau^1,\dots,\tau^M}$ (note that we consider a normalisation constant C a part of each M-fold sum to keep each M-fold sum normalised). This follows since $\Phi_{\rm E}$ and each $\Phi'_{\rm P,\alpha}$ is a linear combination of determinants $\Phi_{\mathbf{r}\sigma}$ respectively permanents $\Phi'_{q_{\alpha}}$, 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 $\Phi'_{\tau^1,...,\tau^M}$ are given by $\varphi'_{\tau^1,...,\tau^M,k}(\mathbf{z}\sigma)$ $= \varphi_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma) \prod_{\alpha=1}^{M} \varphi'_{q_{\alpha},\tau_{k}^{\alpha}}(q_{\alpha} - \frac{\bar{p}_{\alpha}}{\sqrt{N}}).$ Each of the τ^{α} sums in the M-fold sum then runs over all permutations τ^{α} of the sequence (1, ..., N), and acts 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. Note that the M-fold sums usually simplify greatly due to identical orbitals in the $\Phi'_{q_{\alpha}}$ permanents. For example, in the special case that all orbitals in each permanent Φ'_{a} are the same, all the $\Phi'_{\tau^1,\dots,\tau^M}$ become identical, and the M-fold sum reduces to a single determinant of $\varphi'_k(\mathbf{z}\sigma) =$ $\varphi_{{f r}\sigma,k}({f r}\sigma)\prod_{lpha=1}^M \varphi_{q_lpha}'(q_lpha-rac{ar p_lpha}{\sqrt N})$ orbitals as for ground states.⁷ Also note that if we to compute a ground state Φ' use the full $v'_{KS}(\mathbf{z})$ to find the $\varphi'_k(\mathbf{z}\sigma)$, instead of as above use the separation to find $\varphi_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)$ and $\varphi'_{q_{\alpha}}(q_{\alpha})$ individually, we must 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 (as 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 Φ' one has to identify all eigenorbitals $\varphi'(\mathbf{z}\sigma)$ that correspond to a $\varphi_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)\prod_{\alpha=1}^{M}\varphi'_{q_{\alpha},\tau_{k}}(q_{\alpha}-\frac{\bar{p}_{\alpha}}{\sqrt{N}})$ of the given Φ' . Unlike the individual, degenerate, $\Phi'_{\tau^{1},\ldots,\tau^{M}}$ or if not using the right orbitals (e.g., for a ground state), the proper Φ' as said have all the non-interacting physical properties (instead of only $\mathbf{z}_k \sigma_k \leftrightarrow \mathbf{z}_l \sigma_l$ anti-symmetry). Especially, they yield a different expression for $n'(\mathbf{z})$ in

terms of the orbitals where $n'(\mathbf{z})$ always separates. This is another strong argument to use these Φ' in dressed KS for all $\lambda_{\alpha} = 0$, as the exact physical eigenstates Ψ' we are to mimic in this case also exhibit all these properties and separating $n'(\mathbf{z})$. In fact most other Φ' do not lead to a valid KS system as they cannot reproduce the separating $n'(\mathbf{z})$ they here are to recreate (of the all $\lambda_{\alpha} = 0$ ground state Ψ' , or to be general of any all $\lambda_{\alpha} = 0$ eigenstate Ψ').

To generalise this to non-zero λ_{α} also for dressed \hat{H}'_{KS} , we may use the same linear combinations of determinants for the Φ' as we do in standard KS. For example, we may for the Φ' ground state still use a single (or simple linear combination of) determinant(s) of $\varphi'_k(\mathbf{z}\sigma)$ orbitals. Only, as the $v'_{KS}(\mathbf{z})$ no longer separates neither do the orbitals, and Φ' only has $\mathbf{z}_k \sigma_k \leftrightarrow \mathbf{z}_l \sigma_l$ anti-symmetry. To recover the right form if we set all $\lambda_{\alpha} = 0$, 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 $\lambda_{\alpha} =$ 0 correspond to a $\varphi_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)\prod_{\alpha=1}^{M}\varphi'_{q_{\alpha},\tau_{k}^{\alpha}}(q_{\alpha}-\frac{\bar{p}_{\alpha}}{\sqrt{N}})$ of Φ' . Using the Φ' linear combinations of M-fold sums with these orbitals we then recover the right Φ' for all $\lambda_{\alpha} = 0$. As pointed out in the introduction to this Section these Φ' are usually not eigenstates of \hat{H}'_{KS} for $\lambda_{\alpha} \neq 0$ though, except in single determinant cases (or if the determinants only differ in their spin), as the correlation of the $\varphi'(\mathbf{z}\sigma)$ orbitals usually lifts the degeneracy of the determinants. If we insist on using eigenstates of H'_{KS} in dressed KS, these are the single determinants of the Φ' we propose, or linear combinations of such determinants that remain degenerate. Both options differ significantly from the full Φ' we propose, and so break the important equivalences for all $\lambda_{\alpha} = 0$ discussed in the introduction to this Section. We would thus have to mimic states of one form by states of a very different form for all $\lambda_{\alpha} = 0$, and therefore also for other λ_{α} . We would also run into the issues discussed at the end of the last paragraph, which would also persist for other λ_{α} (even if some $v'_{KS}(\mathbf{z})$ is able to recreate $n'(\mathbf{z})$ if all λ_{α} are non-zero it would not give anything sensible). It is beyond our scope to analyse the precise consequences of not using eigenstates for Φ' , but it seems a lessor issue.

For propagation, $\Phi'(t)$ keeps its initial $\mathbf{z}\sigma$ -orbital form if it is a linear combination of determinants (like above), so it stays $\mathbf{z}_k \sigma_k \leftrightarrow \mathbf{z}_l \sigma_l$ anti-symmetric and numerically efficient. The initial state Φ'_0 may have further properties, but these are only kept under evolution using a standard $\hat{H}'_{\mathrm{KS}}(t)$, or a dressed $\hat{H}'_{\mathrm{KS}}(t)$ if all $\lambda_{\alpha}=0$. For example, our examples in the main text are special in the initial states are prepared outside the cavity for $\lambda_{\alpha}=\dot{J}_{\alpha}(t)=0$, so the Φ'_0 have all the non-interacting physical properties, but the $\Phi'(t)$ only keep the $\mathbf{z}_k \sigma_k \leftrightarrow \mathbf{z}_l \sigma_l$ anti-symmetry.

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.

⁷ The actual number of determinants of Φ' still grows fast with the excitation levels, but at least for few modes and low excitations the number of different $\mathbf{z}\sigma$ -orbitals remains feasible. This allows all practical uses expressible directly in terms of the $\mathbf{z}\sigma$ -orbitals in an efficient form. This includes propagation of the $\mathbf{z}\sigma$ -orbitals, but also say density computations, as we may compute the $n'(\mathbf{z})$ of an M-fold sum $\Phi_{\mathbf{r}\sigma}\prod_{\alpha=1}^{M}\Phi'_{q_{\alpha}}$ directly from the $\mathbf{z}\sigma$ -orbitals by $n'(\mathbf{z}) = \left[\sum_{k=1}^{N}|\varphi_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)|^2\right]\prod_{\alpha=1}^{M}\left[\frac{1}{N}\sum_{k_{\alpha}=1}^{N}|\varphi'_{q_{\alpha},k_{\alpha}}(q_{\alpha}-\frac{\bar{p}_{\alpha}}{\sqrt{N}})|^2\right]$ $=\frac{1}{N^{M}}\sum_{k,k_{1},...,k_{M}=1}^{N}\left|\varphi_{\mathbf{r}\sigma,k}(\mathbf{r}\sigma)\prod_{\alpha=1}^{M}\varphi'_{q_{\alpha},k_{\alpha}}(q_{\alpha}-\frac{\bar{p}_{\alpha}}{\sqrt{N}})|^2$, which usually simplifies a lot further due to identical orbitals. The $n'(\mathbf{z})$ of the full Φ' is then a linear combination of the densities of the $N_{\mathbf{r}\sigma}p(Q_{1})...p(Q_{M})$ M-fold sums of Φ' , where $N_{\mathbf{r}\sigma}$ is the number of $\Phi_{\mathbf{r}\sigma}$ in Φ_{E} and $p(Q_{\alpha})$ is the integer partition function of the total excitation level of mode α . This already directly allows for few modes and low excitations, and the many identical orbitals also allow further simplifications here, to allow higher excitations.

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 $0 \le n \le N$ spatial coordinates and a subset $\{p_{\beta}\}\$ of the photon coordinates p_{α} . To evaluate it, one generally has to perform a coordinate transformation: Typically, $\Phi'(t)$ is given in terms of the $q_{\alpha,k}$ -coordinates, and one transforms the p_{β} of \hat{O} to $q_{\beta,k}$, though one may also transform $\Phi'(t)$ to $p_{\alpha,k}$ -coordinates. 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.⁸ 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 $\Phi(t)\chi(t)$, all $O(\mathbf{r}_1,...,\mathbf{r}_n,\{p_\beta\},t)$ expectation values still 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 $J_{\alpha}(t)$. We 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, 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 this system also produces the different forces.

A. Physical Equations of Motion

We consider the physical electron-photon Hamiltonian

$$\hat{H}(t) = \sum_{k=1}^{N} \left[-\frac{1}{2} \nabla_{\mathbf{r}_k}^2 + v(\mathbf{r}_k, t) \right] + \frac{1}{2} \sum_{k \neq l} w(\mathbf{r}_k, \mathbf{r}_l)$$
 (4)

$$+\sum_{\alpha=1}^{M}\bigg[-\tfrac{1}{2}\tfrac{\partial^2}{\partial p_\alpha^2}+\tfrac{1}{2}\Big(\omega_\alpha p_\alpha-\boldsymbol{\lambda}_\alpha\cdot\sum_{k=1}^{N}\mathbf{r}_k\Big)^2\!\!+\tfrac{j_\alpha(t)}{\omega_\alpha}p_\alpha\bigg].$$

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

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

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

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

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

$$\mathbf{F}_{\mathrm{dip}}(\mathbf{r},t) = -\sum_{\alpha=1}^{M} \boldsymbol{\lambda}_{\alpha} \langle \Psi(t) | (\boldsymbol{\lambda}_{\alpha} \cdot \sum_{k=1}^{N} \mathbf{r}_{k}) \hat{n}(\mathbf{r}) | \Psi(t) \rangle = \\ -\sum_{\alpha=1}^{M} \boldsymbol{\lambda}_{\alpha} n(\mathbf{r}t) (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}) - 2 \sum_{\alpha=1}^{M} \boldsymbol{\lambda}_{\alpha} \int d\mathbf{r}' \rho_{2}(\mathbf{r}, \mathbf{r}', t) (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}'), \\ \mathbf{F}_{\mathrm{dip}}^{(1)}(\mathbf{r},t) \qquad \mathbf{F}_{\mathrm{dip}}^{(2)}(\mathbf{r},t)$$

$$\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t) = \sum_{\alpha=1}^{M} \boldsymbol{\lambda}_{\alpha} \langle \Psi(t) | \omega_{\alpha} p_{\alpha} \hat{n}(\mathbf{r}) | \Psi(t) \rangle,$$

are the forces the photons exert on the electron density [2]. Here $\mathbf{F}_{\text{lin}}(\mathbf{r},t)$ is due to the bilinear coupling between the displacement field and the electrons, while $\mathbf{F}_{\text{dip}}(\mathbf{r},t)$ is due to the dipole self-interaction, and balances the bilinear 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]

$$\frac{\partial^2}{\partial t^2} p_{\alpha}(t) = -\omega_{\alpha}^2 p_{\alpha}(t) + \omega_{\alpha} \lambda_{\alpha} \cdot \mathbf{R}(t) - \frac{\dot{J}_{\alpha}(t)}{\omega_{\alpha}},$$

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

B. Dressed Equations of Motion

We consider the dressed electron-photon Hamiltonian

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

⁸ Also note that $n(\mathbf{r},t) = N \sum_{\sigma_1,...,\sigma_N} \int \mathrm{d}^{3(N-1)} r \, \mathrm{d}^{MN} q \, |\Phi'(t)|^2$ yields the same no matter which \mathbf{r}_k one does not integrate over, even if $\Phi'(t)$ has no $\mathbf{r}_k \sigma_k \leftrightarrow \mathbf{r}_l \sigma_l$ anti-symmetry. This follows as $\sum_{\sigma_1,...,\sigma_N} \int \mathrm{d}^{MN} q \, |\Phi'(t)|^2$ has $\mathbf{r}_k \leftrightarrow \mathbf{r}_l$ symmetry by a similar argument as above. The asymmetric definition, the symmetric $n(\mathbf{r},t) = \sum_{k=1}^N \langle \Phi'(t) | \delta(\mathbf{r} - \mathbf{r}_k) | \Phi'(t) \rangle$ and $n(\mathbf{r},t) = \int \mathrm{d}^M q \, n'(\mathbf{z},t)$ therefore all give the same $n(\mathbf{r},t)$. Similarly, for example $p_\alpha(t)$ and $\rho_2(\mathbf{r},\mathbf{r}',t)$ remain uniquely defined. For approximate $\Psi'(t)$, if $\Psi'(t) = \Psi(t)\chi(t)$, $n(\mathbf{r},t)$, $p_\alpha(t)$ and $\rho_2(\mathbf{r},\mathbf{r}',t)$ are for example further the same whether computed using $\Psi'(t)$ or $\Psi(t)$, even if $\Psi'(t)$ lacks $\mathbf{r}_k \sigma_k \leftrightarrow \mathbf{r}_l \sigma_l$ anti-symmetry or $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ symmetry.

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

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

Taking the second time-derivative, we then arrive at

$$\frac{\partial^2}{\partial t^2} n'(\mathbf{z}, t) = \nabla_{\mathbf{z}} \cdot [n'(\mathbf{z}, t) \nabla_{\mathbf{z}} v'(\mathbf{z}, t)]
- \nabla_{\mathbf{z}} \cdot [\mathbf{Q}'_{kin}(\mathbf{z}, t) + \mathbf{Q}'_{int}(\mathbf{z}, t)],$$
(5)

where $\mathbf{Q}'_{\mathrm{kin}}(\mathbf{z},t) = \mathrm{i}\langle \Psi'(t)|[\hat{T}',\hat{\mathbf{j}}'(\mathbf{z})]|\Psi'(t)\rangle$ and $\mathbf{Q}'_{\mathrm{int}}(\mathbf{z},t) = \mathrm{i}\langle \Psi'(t)|[\hat{W}',\hat{\mathbf{j}}'(\mathbf{z})]|\Psi'(t)\rangle = -2\int \mathrm{d}\mathbf{z}'\rho_2'(\mathbf{z},\mathbf{z}',t)\nabla_{\mathbf{z}}w'(\mathbf{z},\mathbf{z}')$ 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. (5) 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 $\dot{J}_{\alpha}(t)$. 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 dressed system, i.e.,

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

Here the dressed stress and photon-matter forces are

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

where we note the $2^{\rm nd}$ term $(N-1){\bf F}_{\rm lin}^{\rm d}({\bf r},t)$ of ${\bf Q}_{\rm int}^{\rm d}({\bf r},t)$ relies on the $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ exchange symmetry to establish that $2\int {\rm d}{\bf r}' {\rm d}^M q \, \rho_2'({\bf z},{\bf z}',t) = (N-1)n'({\bf r},q_1',...,q_M',t).$ 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 $2\sum_{\alpha=1}^{M} \lambda_{\alpha} \int \mathrm{d}^{M}q \mathrm{d}^{3+M}z' \frac{\omega_{\alpha}q'_{\alpha}}{\sqrt{N}} \rho'_{2}(\mathbf{z},\mathbf{z}',t)$, 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,

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

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

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

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

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

In the physical coordinates the mode-resolved Maxwell equations instead originated from the second line of the physical Hamiltonian (4).

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

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

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

This we can also easily confirm by using that for a dressed system corresponding to a physical one $\Psi'(t) = \Psi(t)\chi(t)$, which together with the $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ exchange symmetry allows us to establish the following exact relations¹⁰

$$\begin{aligned} \mathbf{Q}_{\mathrm{kin}}^{\mathrm{d}}(\mathbf{r},t) &= \mathbf{Q}_{\mathrm{kin}}(\mathbf{r},t), \\ \mathbf{Q}_{\mathrm{int}}^{\mathrm{d}}(\mathbf{r},t) &= \mathbf{Q}_{\mathrm{int}}(\mathbf{r},t) + \frac{N-1}{N} \mathbf{F}_{\mathrm{lin}}(\mathbf{r},t) + \mathbf{F}_{\mathrm{dip}}^{(2)}(\mathbf{r},t), \\ \mathbf{F}_{\mathrm{dip}}^{\mathrm{d}}(\mathbf{r},t) &= \mathbf{F}_{\mathrm{dip}}^{(1)}(\mathbf{r},t), \\ \mathbf{F}_{\mathrm{lin}}^{\mathrm{d}}(\mathbf{r},t) &= \frac{1}{N} \mathbf{F}_{\mathrm{lin}}(\mathbf{r},t). \end{aligned}$$

⁹ Remember from Footnote 8 that $n(\mathbf{r},t)$, $\rho_2(\mathbf{r},\mathbf{r}',t)$ and $p_{\alpha}(t)$ are uniquely defined even if $\Psi'(t)$ only has $\mathbf{z}_k\sigma_k\leftrightarrow\mathbf{z}_l\sigma_l$ symmetry, so we need no further properties to use these here.

Note that the physical force expressions also apply even for some non-physical $\Psi'(t)$. In fact the $\mathbf{Q}_{\mathrm{int}}(\mathbf{r},t)$, $\mathbf{F}_{\mathrm{dip}}^{(1)}(\mathbf{r},t)$ and $\mathbf{F}_{\mathrm{dip}}^{(2)}(\mathbf{r},t)$ expressions are useful for any $\Psi'(t)$. $\mathbf{Q}_{\mathrm{kin}}(\mathbf{r},t)$ and $\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$ are only defined given a $\Psi(t)$, so for these we need $\Psi'(t) = \Psi(t)\chi(t)$, in which case one can also compute $n(\mathbf{r},t)$ and $\rho_2(\mathbf{r},\mathbf{r}',t)$ by $\Psi(t)$ instead of $\Psi'(t)$. We further assumed $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ symmetry to obtain the last relation and the second term of $\mathbf{Q}_{\mathrm{int}}^d(\mathbf{r},t)$, though it is possible that $\Psi'(t) = \Psi(t) \prod_{\alpha=1}^M \chi_\alpha(t)$ would suffice instead. Besides these assumptions, all we used to show the four relations are the definitions of $\hat{n}'(\mathbf{z})$, p_α and $\hat{n}(\mathbf{r})$ used for the last relation.

These relations also provide insight into how the forces get recast in the dressed system. For example, they show that the kinetic term is the same in both cases, and that $\mathbf{F}_{\text{lin}}^{\text{d}}(\mathbf{r},t)$ gives $\frac{1}{N}\mathbf{F}_{\text{lin}}(\mathbf{r},t)$, while $\mathbf{Q}_{\text{int}}^{\text{d}}(\mathbf{r},t)$ yields the rest. Further, $\mathbf{F}_{\text{dip}}(\mathbf{r},t)$ comes from the $(\lambda_{\alpha} \cdot \hat{\mathbf{R}})^2$ 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 $\mathbf{F}_{\text{dip}}^{(1)}(\mathbf{r},t)$ both in the physical and the dressed system, while the two-body part provides the remaining $\mathbf{F}_{\text{dip}}^{(2)}(\mathbf{r},t)$, so the dipole self-interaction is indeed treated the same in the two formulations (also in case of a KS calculation).

E. Dressed Kohn-Sham Equations of Motion

We consider the dressed KS Hamiltonian

$$\hat{H}'_{KS}(t) = \sum_{k=1}^{N} \left[-\frac{1}{2} \nabla_{\mathbf{z}_k}^2 + v'(\mathbf{z}_k, t) + v'_{Mxc}(\mathbf{z}_k, t) \right],$$

of a non-interacting auxiliary KS system, that if using a self-consistent time-propagation by design reproduces the physical dressed density $n'[\Psi'_0,v']$ of propagating Ψ'_0 by $v'(\mathbf{z},t)$. To ensure this, the Mxc potential $v'_{\mathrm{Mxc}}[\Psi'_0,\Phi'_0,n']$ is defined as the potential difference $v'_s[\Phi'_0,n']-v[\Psi'_0,n']$ between a non-interacting and interacting system with the same dressed density $n'(\mathbf{z},t)$, since the KS potential $v'_{\mathrm{KS}}[\Psi'_0,\Phi'_0,n',v']=v'+v'_{\mathrm{Mxc}}[\Psi'_0,\Phi'_0,n']$ for the desired dressed density $n'[\Psi'_0,v']$ then reduces to the unique non-interacting potential $v'_s[\Phi'_0,n'[\Psi'_0,v']]$ that reproduces $n'[\Psi'_0,v']$ starting from Φ'_0 . In terms of forces, $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ is given by equating the equations of motion for $n'(\mathbf{z},t)$ for a non-interacting and interacting (5) system,

$$\nabla_{\mathbf{z}} \cdot [n'(\mathbf{z}, t) \nabla_{\mathbf{z}} v'_{\text{Mxc}}(\mathbf{z}, t)] = \nabla_{\mathbf{z}} \cdot [\mathbf{Q}'_{\text{kin,s}}(\mathbf{z}, t) - \mathbf{Q}'_{\text{kin}}(\mathbf{z}, t)] - \nabla_{\mathbf{z}} \cdot \mathbf{Q}'_{\text{int}}(\mathbf{z}, t),$$

and models the force differences (not from the potentials) between the two systems to get the same forces in the two systems. Here $\mathbf{Q}'_{\mathrm{kin,s}}(\mathbf{z},t) = \mathrm{i} \langle \Phi'(t) | [\hat{T}',\hat{\mathbf{j}}'(\mathbf{z})] | \Phi'(t) \rangle$ is the non-interacting dressed momentum-stress forces.

The equation of motion for the dressed KS density is,

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

Here $\mathbf{Q}'_{\mathrm{kin,KS}}(\mathbf{z},t) = \mathrm{i}\langle\Phi'(t)|[\hat{T}',\hat{\mathbf{j}}'(\mathbf{z})]|\Phi'(t)\rangle$ is the dressed KS momentum-stress forces, and equals $\mathbf{Q}'_{\mathrm{kin,s}}(\mathbf{z},t)$, but where we use the KS to indicate it is of a KS system. We mainly think of $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ evaluated at self-consistency, as it usually is in time-dependent theory, but all equations of motion that we write up apply regardless.

Like in Section III C, we may again obtain the equation of motion for $n(\mathbf{r},t)$ by integration. However, this time expressed in terms of 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 $v'_{\text{Mxc}}(\mathbf{z},t)$ term. The equation of motion that we get therefore also hold even for approximate $v'_{\text{Mxc}}(\mathbf{z},t)$,

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

Here the dressed KS stress and photon-matter forces are

$$\begin{aligned} \mathbf{Q}_{\mathrm{kin,KS}}^{\mathrm{d}}(\mathbf{r},t) &= \mathrm{i} \langle \Phi'(t) | [\hat{T},\hat{\mathbf{j}}(\mathbf{r})] | \Phi'(t) \rangle, \\ \mathbf{F}_{\mathrm{dip,KS}}^{\mathrm{d}}(\mathbf{r},t) &= -\sum_{\alpha=1}^{M} \boldsymbol{\lambda}_{\alpha} n(\mathbf{r},t) (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}), \\ \mathbf{F}_{\mathrm{lin,KS}}^{\mathrm{d}}(\mathbf{r},t) &= \sum_{\alpha=1}^{M} \boldsymbol{\lambda}_{\alpha} \int \mathrm{d}^{M} q \frac{\omega_{\alpha} q_{\alpha}}{\sqrt{N}} n'(\mathbf{z},t), \end{aligned}$$

which only differ from the dressed physical $\mathbf{Q}_{\mathrm{kin}}^{\mathrm{d}}(\mathbf{r},t)$, $\mathbf{F}_{\mathrm{dip}}^{\mathrm{d}}(\mathbf{r},t)$ and $\mathbf{F}_{\mathrm{lin}}^{\mathrm{d}}(\mathbf{r},t)$ in that they are based on $\Phi'(t)$. Especially, the two latter equal $\mathbf{F}_{\mathrm{dip}}^{\mathrm{d}}(\mathbf{r},t)$ and $\mathbf{F}_{\mathrm{lin}}^{\mathrm{d}}(\mathbf{r},t)$ up to how well $\Phi'(t)$ reproduces $n(\mathbf{r},t)$ respectively $n'(\mathbf{z},t)$. If we also define $\mathbf{F}_{\mathrm{Mxc}}(\mathbf{r},t) = -\int \mathrm{d}^{M}qn'(\mathbf{z},t)\nabla_{\mathbf{r}}v'_{\mathrm{Mxc}}(\mathbf{z},t)$, we find for the exact self-consistent KS system that

$$\begin{split} \mathbf{F}_{\mathrm{Mxc}}(\mathbf{r},t) &= \mathbf{Q}_{\mathrm{kin}}^{\mathrm{d}}(\mathbf{r},t) - \mathbf{Q}_{\mathrm{kin,KS}}^{\mathrm{d}}(\mathbf{r},t) + \mathbf{Q}_{\mathrm{int}}^{\mathrm{d}}(\mathbf{r},t) \\ &= \mathbf{Q}_{\mathrm{kin}}(\mathbf{r},t) - \mathbf{Q}_{\mathrm{kin,KS}}^{\mathrm{d}}(\mathbf{r},t) \\ &+ \mathbf{Q}_{\mathrm{int}}(\mathbf{r},t) + \frac{N-1}{N} \mathbf{F}_{\mathrm{lin}}(\mathbf{r},t) + \mathbf{F}_{\mathrm{dip}}^{(2)}(\mathbf{r},t), \\ \mathbf{F}_{\mathrm{dip,KS}}^{\mathrm{d}}(\mathbf{r},t) &= \mathbf{F}_{\mathrm{dip}}^{\mathrm{d}}(\mathbf{r},t) = \mathbf{F}_{\mathrm{dip}}^{(1)}(\mathbf{r},t), \\ \mathbf{F}_{\mathrm{lin,KS}}^{\mathrm{d}}(\mathbf{r},t) &= \mathbf{F}_{\mathrm{lin}}^{\mathrm{d}}(\mathbf{r},t) = \frac{1}{N} \mathbf{F}_{\mathrm{lin}}(\mathbf{r},t), \end{split}$$

so $\mathbf{F}_{\mathrm{Mxc}}(\mathbf{r},t) + \mathbf{Q}_{\mathrm{kin,KS}}^{\mathrm{d}}(\mathbf{r},t) + \mathbf{F}_{\mathrm{dip,KS}}^{\mathrm{d}}(\mathbf{r},t) + \mathbf{F}_{\mathrm{lin,KS}}^{\mathrm{d}}(\mathbf{r},t) = \mathbf{Q}_{\mathrm{kin}}(\mathbf{r},t) + \mathbf{Q}_{\mathrm{int}}(\mathbf{r},t) + \mathbf{F}_{\mathrm{dip}}(\mathbf{r},t) + \mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$, and we get the same equation of motion for $n(\mathbf{r},t)$ as in the physical system. We automatically get $\mathbf{Q}_{\mathrm{kin,KS}}^{\mathrm{d}}(\mathbf{r},t) + \mathbf{F}_{\mathrm{dip}}^{(1)}(\mathbf{r},t) + \frac{1}{N}\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$ (and the forces from $v(\mathbf{r},t)$), while $\mathbf{F}_{\mathrm{Mxc}}(\mathbf{r},t)$ then has to model the rest, as discussed in the main text.

Likewise we can again obtain the equation of motion for $p_{\alpha}(t)$, which also holds even for approximate $v'_{\text{Mxc}}(\mathbf{z},t)$, by multiplying the equation of motion for $n'(\mathbf{z},t)$ by $\frac{q_{\alpha}}{\sqrt{N}}$, and integrating this over all \mathbf{z} coordinates (again we may reuse the results from Section III C),

$$\frac{\partial^2}{\partial t^2} p_{\alpha}(t) = -\omega_{\alpha}^2 p_{\alpha}(t) + \frac{\omega_{\alpha}}{N} \boldsymbol{\lambda}_{\alpha} \cdot \mathbf{R}(t) - \frac{j_{\alpha}(t)}{\omega_{\alpha}} - \frac{1}{\sqrt{N}} \int d\mathbf{z} \, n'(\mathbf{z}, t) \frac{\partial}{\partial q_{\alpha}} v'_{\text{Mxc}}(\mathbf{z}, t).$$

To get the exact mode-resolved Maxwell equations the $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ term has to contribute $\frac{N-1}{N}\omega_{\alpha}\boldsymbol{\lambda}_{\alpha}\cdot\mathbf{R}(t)$. This is the contribution we get from $w'(\mathbf{z},\mathbf{z}')$ in the interacting system. One may separate $-\frac{N-1}{N}\sum_{\alpha=1}^{M}\frac{\omega_{\alpha}q_{\alpha}}{\sqrt{N}}\boldsymbol{\lambda}_{\alpha}\cdot\mathbf{R}(t)$ out of $v'_{\mathrm{Mxc}}(\mathbf{z},t)$ for this (which is a $\frac{N-1}{N}$ part of an identical term in the standard KS mean-field approximation).

To reproduce the exact $n'(\mathbf{z}, t)$ we of course need to design $v'_{\text{Mxc}}(\mathbf{z}, t)$ 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 $n'(\mathbf{z},t)$. This compensation is mainly needed to model $\mathbf{F}_{\text{lin}}(\mathbf{r},t)$, as it is the one term in the equations of motion for $n(\mathbf{r},t)$ and $p_{\alpha}(t)$ that strongly depends on the electron-photon correlations. This force can be highly sensitive though. This is a good reason to aim to get $n'(\mathbf{z},t)$ right. However, as only one $v'_{\text{Mxc}}(\mathbf{z},t)$ get $n'(\mathbf{z},t)$ right, while many get $n(\mathbf{r},t)$ and $p_{\alpha}(t)$ right, the latter may still be easier.

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

We consider the standard cavity QEKS Hamiltonian [2]

$$\begin{split} \hat{H}_{\mathrm{KS}}(t) &= \sum_{k=1}^{N} \left\{ -\frac{1}{2} \nabla_{\mathbf{r}_{k}}^{2} + v(\mathbf{r}_{k}, t) + v_{\mathrm{mxc}}(\mathbf{r}_{k}, t) \right. \\ &+ \sum_{\alpha=1}^{M} [\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{R}(t) - \omega_{\alpha} p_{\alpha}(t)] \boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}_{k} \right\} \\ &+ \sum_{\alpha=1}^{M} \left[-\frac{1}{2} \frac{\partial^{2}}{\partial p_{\alpha}^{2}} + \frac{1}{2} \omega_{\alpha}^{2} p_{\alpha}^{2} - \omega_{\alpha} p_{\alpha} \boldsymbol{\lambda}_{\alpha} \cdot \mathbf{R}(t) + \frac{j_{\alpha}(t)}{\omega_{\alpha}} p_{\alpha} \right], \end{split}$$

of a non-interacting, non-correlated auxiliary KS system that at self-consistency shares the same density $n(\mathbf{r},t)$ and photon-displacements $p_{\alpha}(t)$ as the physical system. To ensure this, $\hat{H}_{\mathrm{KS}}(t)$ incorporates an explicit mean-field contribution, where $\mathbf{R}(t)$ and $p_{\alpha}(t)$ are the expectation values of $\hat{\mathbf{R}}$ and p_{α} , and $v_{\mathrm{mxc}}(\mathbf{r},t)$ denotes the remaining exchange-correlation potential. For the initial state Φ_0 , we in general use a linear combination of the eigenstates $\Phi_{\mathrm{E}} \prod_{\alpha=1}^{M} \varphi_{p_{\alpha}}(p_{\alpha})$ of the KS Hamiltonian \hat{H}_{KS} (especially, we generally use the ground state if Ψ_0 is a ground state). Here the electronic eigenstate Φ_{E} is in general a weighted sum of Slater determinants $\Phi_{\mathbf{r}\sigma}$ of orbitals with spatial part $\varphi_{\mathbf{r},k}(\mathbf{r},t)$. This leads to the single particle equations

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

and the force equations,

$$\begin{split} \frac{\partial^2}{\partial t^2} n(\mathbf{r},t) &= \nabla_{\mathbf{r}} \cdot \left\{ n(\mathbf{r},t) \nabla_{\mathbf{r}} [v(\mathbf{r},t) + v_{\text{mxc}}(\mathbf{r},t)] \right\} \\ &- \nabla_{\mathbf{r}} \cdot \left\{ n(\mathbf{r},t) \sum_{\alpha=1}^{M} \boldsymbol{\lambda}_{\alpha} [\omega_{\alpha} p_{\alpha}(t) - \boldsymbol{\lambda}_{\alpha} \cdot \mathbf{R}(t)] \right\} \\ &- \nabla_{\mathbf{r}} \cdot \mathbf{Q}_{\text{kin,KS}}(\mathbf{r},t), \\ \frac{\partial^2}{\partial t^2} p_{\alpha}(t) &= -\omega_{\alpha}^2 p_{\alpha}(t) + \omega_{\alpha} \boldsymbol{\lambda}_{\alpha} \cdot \mathbf{R}(t) - \frac{j_{\alpha}(t)}{\omega_{\alpha}}, \end{split}$$

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. Here $\mathbf{Q}_{\mathrm{kin,KS}}(\mathbf{r},t)=\mathrm{i}\langle\Phi(t)|[\hat{T},\hat{\mathbf{j}}(\mathbf{r})]|\Phi(t)\rangle$ is the standard KS momentum-stress forces. Note that one may solve the Maxwell equations analytically, $p_{\alpha}(t)=p_{\alpha,0}\cos(\omega_{\alpha}t)+\frac{\dot{p}_{\alpha,0}}{\omega_{\alpha}}\sin(\omega_{\alpha}t)+\int_{0}^{t}\sin[\omega_{\alpha}(t-t')][\boldsymbol{\lambda}_{\alpha}\cdot\mathbf{R}(t')-\frac{\dot{j}_{\alpha}(t')}{\omega_{\alpha}^{2}}]\mathrm{d}t'$, and substitute the results into the single particle equations for $\varphi_{\mathbf{r},k}(\mathbf{r},t)$. This usually saves one to compute $\varphi_{p_{\alpha}}(p_{\alpha},t)$.

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 H_{aux} to $H_{\text{KS}}(t)$ and switch coordinates to with $v'_{\text{KS}}(\mathbf{z}, t) = v(\mathbf{r}, t) + v_{\text{mxc}}(\mathbf{r}, t) + \sum_{\alpha=1}^{M} \left[\frac{1}{2} \omega_{\alpha}^2 q_{\alpha}^2 - \frac{\omega_{\alpha}}{\sqrt{N}} q_{\alpha} \boldsymbol{\lambda}_{\alpha} \cdot \mathbf{R}(t) + \frac{j_{\alpha}(t)}{\sqrt{N}\omega_{\alpha}} q_{\alpha} \right]$. We get no \hat{W}'_{KS} , as this is a real intersection dropped KS Hamiltonian. Note so this is a non-interacting dressed KS Hamiltonian. Note that $v_{\text{mxc}}(\mathbf{r},t)$ depends only on \mathbf{r} , as usual in standard KS, while the dressed $v'_{\text{Mxc}}(\mathbf{z},t)$ depends on the full \mathbf{z} . By noting that $\hat{H}_{\mathrm{KS}}(t) = \hat{H}_{\mathrm{KS,E}}(t) + \sum_{\alpha=1}^{M} \hat{H}_{p_{\alpha}}(p_{\alpha} - \bar{p}_{\alpha}(t))$ with $\bar{p}_{\alpha}(t) = -\frac{j_{\alpha}(t)}{\omega_{\alpha}^{3}} + \frac{\lambda_{\alpha} \cdot \mathbf{R}(t)}{\omega_{\alpha}}$, we also see that $\hat{H}'_{\mathrm{KS}}(t) = \hat{H}_{\mathrm{KS,E}}(t) + \sum_{\alpha=1}^{M} \hat{H}'_{\mathrm{KS,P,\alpha}}(t)$. Hence the eigenstates take the form discussed in Section II.P. For the contribution of the form discussed in Section II.P. For the contribution of the form discussed in Section II.P. For the contribution of the form discussed in Section II.P. For the contribution of the form discussed in Section II.P. For the contribution of the form discussed in Section II.P. For the contribution of the form discussed in Section II.P. For the contribution of the form discussed in Section II.P. For the contribution of the form discussed in Section II.P. For the contribution of the form discussed in Section II.P. For the contribution of the form discussed in Section II.P. For the contribution of the form discussed in Section II.P. For the contribution II.P. For t the form discussed in Section II B. For the initial state we use $\Phi'_0 = \Phi_0 \chi_0$ (χ_0 is usually the $q_{\alpha,k} \leftrightarrow q_{\alpha,l}$ symmetric ground state of \hat{H}_{aux} , but may be any linear combination of eigenstates). This is a linear combination of eigenstates $\Phi_{\rm E} \prod_{\alpha=1}^{M} \varphi_{p_{\alpha}}(p_{\alpha} - \bar{p}_{\alpha}) \chi_{\alpha} \text{ of } \hat{H}'_{\rm KS} = \hat{H}_{\rm KS} + \hat{H}_{\rm aux} \text{ (usually }$ just the ground state), and so one may also construct Φ'_0 directly using H'_{KS} instead of going through Φ_0 . Finally, we note that $\Phi'(t) = \Phi(t)\chi(t)$, and standard KS therefore produces the $n'(\mathbf{z},t)$ of this $\Phi'(t)$.

As a side remark, for the two electrons in a singlet state coupled with one mode examples of the main text, the standard mean-field and Mx approximations to $v_{\rm mxc}({\bf r},t)$ are given by

$$v_{\text{MF}}(\mathbf{r}, t) = \frac{1}{2} \int d\mathbf{r}' \, n(\mathbf{r}', t) w(\mathbf{r}, \mathbf{r}'),$$

$$v_{\text{Mx}}(\mathbf{r}, t) = \frac{1}{2} \int d\mathbf{r}' \, n(\mathbf{r}', t) w(\mathbf{r}, \mathbf{r}')$$

$$+ \frac{1}{2} (\boldsymbol{\lambda} \cdot \mathbf{r})^2 - \frac{1}{2} [\boldsymbol{\lambda} \cdot \mathbf{R}(t)] (\boldsymbol{\lambda} \cdot \mathbf{r}),$$

as our naming convention refers only to the light-matter interaction (while the Coulomb interaction in both cases is treated on an exact exchange level). The Mx approximation again approximates the forces by their KS values.

In conclusion, standard cavity QEKS is a special case of our dressed KS, but 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. SCALED APPROXIMATION

In this Section we discuss a first simple approximation for the Mxc potential that makes use of the force relations that we established in Section III. Specifically, we redistribute the $\frac{N-1}{N}\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$ forces originating from $w'(\mathbf{z},\mathbf{z}')$ to $v'_{\mathrm{KS}}(\mathbf{z},t)$, by scaling by N the term in $v'_{\mathrm{KS}}(\mathbf{z},t)$ responsible for $\frac{1}{N}\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$. This simple idea ensures that the dressed KS system trivially shares the exact same force expression for $\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$ as the original system.

For the dressed system,

$$v'(\mathbf{z},t) = v(\mathbf{r},t) + \sum_{\alpha=1}^{M} \left[\frac{1}{2} \omega_{\alpha}^{2} q_{\alpha}^{2} \underbrace{-\frac{\omega_{\alpha}}{\sqrt{N}} q_{\alpha}(\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r})}_{\text{Tin}} + \frac{1}{2} (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r})^{2} + \frac{j_{\alpha}(t)q_{\alpha}}{\sqrt{N}\omega_{\alpha}} \right],$$

$$+ \frac{1}{2} (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r})^{2} + \frac{j_{\alpha}(t)q_{\alpha}}{\sqrt{N}\omega_{\alpha}} \right],$$

$$w'(\mathbf{z}, \mathbf{z}') = w(\mathbf{r}, \mathbf{r}') + \sum_{\alpha=1}^{M} \left[(\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r})(\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}') - \frac{\omega_{\alpha}}{\sqrt{N}} q_{\alpha}'(\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}') - \frac{\omega_{\alpha}}{\sqrt{N}} q_{\alpha}'(\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}) \right],$$

$$\to (N-1)\mathbf{F}_{\text{lin}}^{d} = \frac{N-1}{N} \mathbf{F}_{\text{lin}} & \frac{N-1}{N} \omega_{\alpha} \boldsymbol{\lambda}_{\alpha} \cdot \mathbf{R}(t)$$

already a fraction $\frac{1}{N}$ of $\mathbf{F}_{\text{lin}}(\mathbf{r},t)$, and of the $\omega_{\alpha} \boldsymbol{\lambda}_{\alpha} \cdot \mathbf{R}(t)$ of each mode-resolved Maxwell equation, is generated by the dressed potential $v'(\mathbf{z},t)$, while the remaining parts $\frac{N-1}{N}$ are generated by $w'(\mathbf{z},\mathbf{z}')$. In our dressed KS scheme those interactions have to be mimicked by the mean-field exchange correlation potential $v'_{\text{Mxc}}(\mathbf{z},t)$.

An interesting idea to do this is to first redistribute the $\frac{N-1}{N}\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$ forces originating from $w'(\mathbf{z},\mathbf{z}')$ to $v'(\mathbf{z},t)$ by scaling the corresponding part $-\frac{\omega_{\alpha}}{\sqrt{N}}q_{\alpha}(\boldsymbol{\lambda}_{\alpha}\cdot\mathbf{r})$ of $v'(\mathbf{z},t)$ by N. That is, we consider a $\tilde{H}'(t)$ system with,

$$\tilde{v}'(\mathbf{z},t) = v(\mathbf{r},t) + \sum_{\alpha=1}^{M} \left[\frac{1}{2} \omega_{\alpha}^{2} q_{\alpha}^{2} \underbrace{-\sqrt{N} \omega_{\alpha} q_{\alpha} (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r})}_{\rightarrow \mathbf{F}_{\text{lin}} \& \omega_{\alpha} \boldsymbol{\lambda}_{\alpha} \cdot \mathbf{R}(t)} + \frac{1}{2} (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r})^{2} + \frac{j_{\alpha}(t) q_{\alpha}}{\sqrt{N} \omega_{\alpha}} \right] + \tilde{v}'_{\text{Mxc}}(\mathbf{z},t),$$

$$\tilde{w}'(\mathbf{z}, \mathbf{z}') = w(\mathbf{r}, \mathbf{r}') + \sum_{\alpha=1}^{M} (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}) (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}').$$

Even for $\tilde{v}'_{\text{Mxc}}(\mathbf{z},t) = 0$, this system shares the same total force expression for $n(\mathbf{r},t)$ as the original physical system

with the minor redistribution that

$$\tilde{\mathbf{Q}}_{\mathrm{int}}^{\mathrm{d}}(\mathbf{r},t) = -2 \int d\mathbf{r}' \rho_{2}(\mathbf{r},\mathbf{r}',t) \nabla_{\mathbf{r}} w(\mathbf{r},\mathbf{r}')$$

$$-2 \sum_{\alpha=1}^{M} \boldsymbol{\lambda}_{\alpha} \int d\mathbf{r}' \rho_{2}(\mathbf{r},\mathbf{r}',t) (\boldsymbol{\lambda}_{\alpha} \cdot \mathbf{r}'),$$

$$\tilde{\mathbf{F}}_{\mathrm{lin}}^{\mathrm{d}}(\mathbf{r},t) = \underbrace{N}_{\alpha} \sum_{\alpha=1}^{M} \boldsymbol{\lambda}_{\alpha} \int d^{M} q \frac{\omega_{\alpha} q_{\alpha}}{\sqrt{N}} n'(\mathbf{z},t).$$

Also the mode-resolved Maxwell equations stay identical

$$\partial_t^2 p_{\alpha}(t) = -\omega_{\alpha}^2 p_{\alpha}(t) + \omega_{\alpha} \lambda_{\alpha} \cdot \mathbf{R}(t) - \frac{j_{\alpha}(t)}{\omega_{\alpha}},$$

with the only redistribution that now all of $\omega_{\alpha} \lambda_{\alpha} \cdot \mathbf{R}(t)$ originates from $\tilde{v}'(\mathbf{z},t)$ instead of $\frac{1}{N}$ from $v'(\mathbf{z},t)$ and $\frac{N-1}{N}$ from $w'(\mathbf{z}, \mathbf{z}')$. That is, the full RHS now originates from $\tilde{v}'(\mathbf{z},t)$ alone. To reproduce $n'(\mathbf{z},t)$ we still need the right $\tilde{v}'_{\rm Mxc}(\mathbf{z},t)$ though (i.e., $\tilde{H}'(t)$ is a KS system). Without it, the underlying system solved H'(t), which enacts forces $\tilde{\mathbf{F}}'(\mathbf{z},t)$ on the dressed density $n'(\mathbf{z},t)$, will still exhibit deviating forces from the original system $\hat{H}'(t)$. That is, $\Psi'(t)$ still differs from $\Psi'(t)$, leading to different forces even for the physical observables $n(\mathbf{r},t)$ and $p_{\alpha}(t)$ despite the force expressions are now exact. 11 This is especially so as this idea has not been designed with $n'(\mathbf{z}, t)$ in mind. Indeed, $\mathbf{F}_{lin}(\mathbf{r},t)$ can be very sensitive to this, since it depends strongly on the electron-photon correlations in $n'(\mathbf{z},t)$, which are treated differently in the H'(t) system. Hence we cannot expect H'(t) to perform well in general without $\tilde{v}'_{\text{Mxc}}(\mathbf{z},t)$. However, it can still be very precise for specific domains of applicability.

In practise, this all leads us to rewrite the $v'_{KS}(\mathbf{z},t) = v'(\mathbf{z},t) + v'_{Mxc}(\mathbf{z},t)$ of the usual dressed KS system as

$$v'_{KS}(\mathbf{z},t) = \bar{v}'(\mathbf{z},t) + v'_{SMXC}(\mathbf{z},t),$$

where $\bar{v}'(\mathbf{z},t)$ is shorthand for $\tilde{v}'(\mathbf{z},t)$ without $\tilde{v}'_{\mathrm{Mxc}}(\mathbf{z},t)$. This generates the exact force expression for $\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$ and for the full mode resolved Maxwell equations even for $v'_{\mathrm{sMxc}}(\mathbf{z},t)=0$, while $v'_{\mathrm{sMxc}}(\mathbf{z},t)=v'_{\mathrm{Mxc}}(\mathbf{z},t)-\frac{1-N}{\sqrt{N}}\omega_{\alpha}q_{\alpha}(\boldsymbol{\lambda}_{\alpha}\cdot\mathbf{r})$ represents the remaining Mxc potential after scaling (i.e., it generates the other forces and further corrections to the bilinear forces, e.g., $\tilde{v}'_{\mathrm{Mxc}}(\mathbf{z},t)$). If we neglect $\tilde{v}'_{\mathrm{Mxc}}(\mathbf{z},t)$, and approximate the contribution from $\tilde{w}(\mathbf{z},\mathbf{z}')$ by its KS value, we get the sMx approximation,

$$\nabla_{\mathbf{z}} \cdot [n'(\mathbf{z}, t) \nabla_{\mathbf{z}} v'_{\text{sMx}}(\mathbf{z}, t)] = -\nabla_{\mathbf{z}} \cdot i \langle \Phi'(t) | [\tilde{W}', \hat{\mathbf{j}}'(\mathbf{z})] | \Phi'(t) \rangle.$$

This is the scaled analogue of the Mx approximation, and differs only in the bilinear forces here are treated by redistribution instead of using the KS values.

¹¹ $\tilde{H}'(t)$ also breaks all the physical symmetries, however, the $n(\mathbf{r},t)$ and $p_{\alpha}(t)$ force expressions still apply as is, as the one expression that relied on these properties for \hat{W}' is no longer present for \tilde{W}' .

We now study how the sMx approximation performs for the two examples in the main text (compared with the Mx results presented there). Note that in both cases, i.e., for two particles in a singlet state coupled to one mode with no external current J(t) = 0 the sMx approximation reduces to

$$\bar{v}'(\mathbf{r}, q, t) = v(\mathbf{r}, t) + \frac{1}{2}\omega^2 q^2 - \sqrt{2}\omega q(\boldsymbol{\lambda} \cdot \mathbf{r}) + \frac{1}{2}(\boldsymbol{\lambda} \cdot \mathbf{r})^2,$$

$$v'_{\text{sMx}}(\mathbf{r}, q, t) = \frac{1}{2} \int d^3 \mathbf{r}' \, n(\mathbf{r}', t) w(\mathbf{r}, \mathbf{r}') + \frac{1}{2} [\boldsymbol{\lambda} \cdot \mathbf{R}(t)] (\boldsymbol{\lambda} \cdot \mathbf{r}).$$

Also note that $v'_{\text{sMx}}(\mathbf{z},t)$ is simply $v'_{\text{Mx}}(\mathbf{z},t)$ without the two bilinear coupling terms.

We first compare our new sMx results with the previously presented Mx results for the two-site case in Fig. 2.

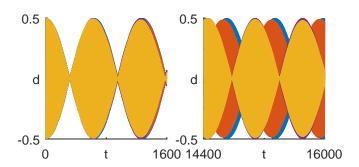


FIG. 2. (color online) The exact (orange), dressed sMx (lilac), dressed Mx (red) and standard Mx (blue) dipole moment of two electrons on two sites coupled to one mode.

Dressed sMx (lilac) improves the accuracy for the given time-period to almost perfect agreement, and greatly outperforms both dressed Mx (red) and standard Mx (blue). However, we should note that if one for example increases λ to 0.5 (not shown), where we only looked at t=[0,100], dressed Mx actually slightly outperforms dressed sMx, but both still perform very decent.

Let us now address the example of spontaneous emission for the one-dimensional helium model in Fig. 3.

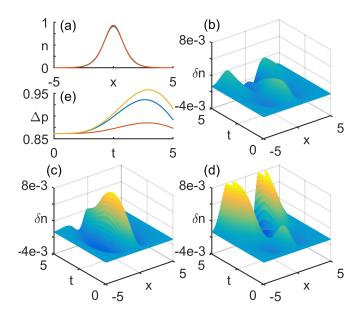


FIG. 3. (color online) (a) The exact (blue) and Mx (red) ground-state densities of the bare $(\lambda=0)$ one-dimensional helium model. The corresponding changes in the (b) exact (c) dressed Mx and (d) dressed sMx density, $\delta n(x,t) = n(x,t) - n_0(x)$, when placed inside a cavity, and (e) the exact (blue), dressed Mx (red) and dressed sMx (orange) field fluctuations.

Dressed sMx (orange and (d)) improves the photonfield fluctuations, and the density is better at short times but worsens for longer times compared to dressed Mx (red and (c)). The density improvement for short times relies on error cancellation though. That is, we only have a density change due to coupling with the photon mode, so the change is mainly due to $\mathbf{F}_{\text{lin}}(\mathbf{r},t)$ and $\mathbf{F}_{\text{dip}}(\mathbf{r},t)$. The errors in these are in opposite directions, so while the $\mathbf{F}_{\mathrm{lin}}(\mathbf{r},t)$ errors are here larger for dressed sMx than dressed Mx, there is still an improvement for short times where the $\mathbf{F}_{\text{dip}}(\mathbf{r},t)$ errors dominate. To understand this case better, an analysis of the individual contributions, and their approximations (and how they work together), is required. We focus in the following on the bilinear coupling, since essentially only this part of H(t) is treated differently in dressed KS than standard KS, and it is also the difference between our dressed sMx and dressed Mx. Here the H'(t) system is very useful, since it lets us study the effect of approximating the bilinear coupling alone (as the other parts are the exact ones). In the following, we therefore compare exact results (obtained using $\hat{H}(t)$) with sM̃x (using $\tilde{H}'(t)$ with $\tilde{v}'_{\rm Mxc}(\mathbf{z},t)=0$) and M̃x (using $\tilde{H}'(t)$ but with the Mx approximation for the bilinear part instead of the scaled potential, i.e., with $-\sqrt{2}\omega q(\boldsymbol{\lambda}\cdot\mathbf{r})$ + $\tilde{v}_{\mathrm{Mxc}}'(\mathbf{z},t) = -\tfrac{\omega}{\sqrt{2}}q(\boldsymbol{\lambda}\cdot\mathbf{r}) - \tfrac{\omega}{2}p(t)(\boldsymbol{\lambda}\cdot\mathbf{r}) - \tfrac{\omega}{2\sqrt{2}}q\boldsymbol{\lambda}\cdot\mathbf{R}(t), \text{ where }$ the $v'_{Mx}(\mathbf{z},t)$ part here fails to contribute by symmetry, i.e., $p(t) = \mathbf{R}(t) = 0$). For convenience we further change to ground state KS, as the physics remains the same.

We start by studying the effect of the bilinear coupling alone. That is, we set $w(\mathbf{r}, \mathbf{r}')$ and the quadratic coupling (all λ^2 terms) to zero for both the exact and \tilde{H}' systems.

This leads to Fig. 4 (a), where the shown change in the ground state density is due to the bilinear coupling alone.

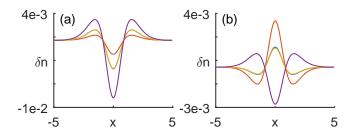


FIG. 4. (color online) The changes in the exact (blue, hidden behind orange), $\tilde{\text{Mx}}$ (red), $\sqrt{\tilde{\text{s}}}\tilde{\text{Mx}}$ (orange) and $\tilde{\text{s}}\tilde{\text{Mx}}$ (lilac) ground state density, $\delta n(x) = n_{\lambda}(x) - n_{\lambda=0}(x)$, of a one-dimensional helium model (a) without w(x,x') and λ^2 terms and (b) without w(x,x').

Here sMx (lilac) gives twice the exact density change (blue, hidden behind orange), while Mx (red) only gives half (as the Mx potential fails to contribute). Hence we need further corrections $\tilde{v}'_{\text{Mxc}}(\mathbf{z})$. We can reproduce the density very well by scaling with \sqrt{N} instead of N though (i.e., using $-\omega q(\boldsymbol{\lambda} \cdot \mathbf{r})$ instead of $-\sqrt{2}\omega q(\boldsymbol{\lambda} \cdot \mathbf{r})$ in $\tilde{v}'(\mathbf{z})$). We call this $\sqrt{s}Mx$ (orange), which may have a domain of applicability to the extent it can be generalised to more electrons and with a further correction asymmetric cases. The change in the exact field fluctuations 0.0102 (0.7091) minus 0.6989 at $\lambda = 0$) is reproduced very accurately by sMx 0.0103, while Mx only yields a quarter of the change 0.0025, and \sqrt{s} Mx half the change 0.0050 (all vs 0.6989). So with scaling alone there is in this case a trade-off where we can either get the density $(\sqrt{s}Mx)$ or field fluctuations (sMx) right, but not both, this requires some $\tilde{v}'_{\text{Mxc}}(\mathbf{z})$.

If we include the λ^2 terms again we get Fig. 4 (b). Here (at least for this coupling strength) the density changes almost equal the sum of the density change for quadratic alone (not shown), and for bilinear alone. Hence we can in this case consider these two approximations separately, and $\sqrt{s} Mx$ together with a good approximation for the quadratic terms will yield excellent densities. Note how the s Mx density change is opposite of what it should be; it is essential to get the right scale to balance the effect of the bilinear and quadratic couplings against each other. The field fluctuations are barely changed by including the λ^2 terms (all changes are reduced but at most by 0.0004), so they are still reproduced very accurately by s Mx.

Including $w(\mathbf{r}, \mathbf{r}')$ again (but again no λ^2 terms at first) we arrive at Fig. 5 (a), where the density change is again only due to the bilinear coupling.

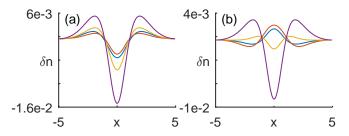


FIG. 5. (color online) The changes in the exact (blue), $\tilde{\text{Mx}}$ (red), $\sqrt{\tilde{\text{s}}}$ Mx (orange) and $\tilde{\text{s}}$ Mx (lilac) ground state density, $\delta n(x) = n_{\lambda}(x) - n_{\lambda=0}(x)$, of a one-dimensional helium model (a) without λ^2 terms and (b) with all terms.

Here regular $\tilde{M}x$ (red) outperforms $\sqrt{s}Mx$ (orange), which now gives a too strong change, and sMx (lilac), which is now even more too strong. This effect of $w(\mathbf{r}, \mathbf{r}')$ on the effect of the bilinear coupling limits how good results we can expect using any fixed scaling on its own (e.g., $\sqrt{s}Mx$ or sMx). We need some $\tilde{v}'_{Mxc}(\mathbf{z})$. Especially, if we include the λ^2 terms again in Fig. 5 (b), it again just adds its own change without affecting the other changes much but now only Mx yields a qualitatively right density change, clearly showing the issue in using wrong scaling. Figure 5 (a) is also a clear example of that while $v'(\mathbf{z})$ gives us $\frac{1}{N}$ of the force expression for $\mathbf{F}_{\text{lin}}(\mathbf{r})$, it only gives us $\frac{1}{N}$ of the force if we use the exact $v'_{\text{Mxc}}(\mathbf{z})$. In this case we then just happen to get more of the force, so Mx (for which the only contribution to $\mathbf{F}_{lin}(\mathbf{r})$ in this case is that from $v'(\mathbf{z})$ is closer to the exact than one would expect. The change in the exact field fluctuations 0.0183 (0.8797) minus 0.8615 for $\lambda = 0$) is now also in between sMx 0.0255and $\sqrt{s}Mx$ 0.0121, so this picture is also less clear here ($\tilde{M}x$ gives 0.0059, all vs 0.8615). The field fluctuations are still barely changed by including the λ^2 terms though (all changes are again reduced and at most by 0.0012).

Finally, we show that we get the same density changes from the bilinear coupling when using KS also for $w(\mathbf{r}, \mathbf{r}')$, i.e., using \hat{H}'_{KS} instead of \tilde{H}' , though it treats the electron correlations differently. Using exact exchange for $w(\mathbf{r}, \mathbf{r}')$, the bare KS and exact ground state differ. However, the density change compared to the bare density is still only due to the bilinear coupling (as we include no λ^2 terms), and in Fig. 6 (a) we confirm that the dressed Mx, $\sqrt{\mathrm{sMx}}$ and sMx density changes are nearly the same as using \tilde{H}' (Fig. 5 (a)).

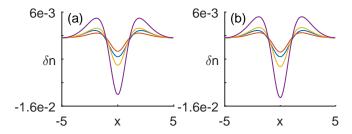


FIG. 6. (color online) (a) The changes in the exact (blue), dressed Mx (red), dressed \sqrt{s} Mx (orange) and dressed sMx (lilac) ground state density, $\delta n(x) = n_{\lambda}(x) - n_{\lambda=0}(x)$, of a one-dimensional helium model without λ^2 and (b) using an improved approximation for the interaction w(x, x').

This remains true also using more precise approximations for $w(\mathbf{r}, \mathbf{r}')$ such as $v'_{KS}(\mathbf{z}) = \bar{v}'(\mathbf{z}) + v'_{sMxc,bare}(\mathbf{z}) + \frac{1}{2} \int d^3\mathbf{r}' \left[n(\mathbf{r}') - n_{bare}(\mathbf{r}') \right] w(\mathbf{r}, \mathbf{r}')$, which uses the exact bare sMxc potential (so the bare densities now agree) added the exact exchange backreaction to the density change. The corresponding density changes in Fig. 6 (b) are even closer to Fig. 5 (a) than in Fig. 6 (a).

In conclusion, we cannot expect to find a scaling that works well on its own in general, especially given the dependence on $w(\mathbf{r}, \mathbf{r}')$, without further modifications. However, scaling may still be very accurate in special cases, as seen in our two-site case for the dipole moment, or field fluctuations of our one-dimensional helium model. Whether the \sqrt{s} Mx approximation can be generalised to

more electrons, and if it has an area of applicability when $w(\mathbf{r}, \mathbf{r}') = 0$, for similar cases, remains to be seen. Clearly there is still much to investigate about scaling in general, for example also by studying the $n'(\mathbf{z}, t)$ equation of motion.

VI. OUTLOOK

In this supplemental material we have provided many details of the properties of the dressed physical and KS systems. We have further highlighted how the dressed KS construction reproduces the exact dressed 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 QEKS 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 the scaled approximation and all the relations between the physical, dressed and KS forces is the subject of ongoing work to find more accurate functionals for dressed QEKS, standard QEKS as well as quantumelectrodynamical reduced density matrix functional theory [6].

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