Supplementary Material for "Exact time-dependent density functional theory for non-perturbative dynamics of helium atom"

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I. NUMERICAL DETAILS

In order to solve the equation,

$$\nabla \cdot (n(\mathbf{r},t)\nabla\alpha(\mathbf{r},t)) = -\frac{\partial}{\partial t} n(\mathbf{r},t) \tag{1}$$

we construct the explicit matrix representation of the operator $\nabla\cdot(n(\mathbf{r},t)\nabla\alpha(\mathbf{r},t))$ subject to the boundary conditions,

$$\alpha(\mathbf{r}\to\infty,t) = 0 \quad \text{and} \quad \frac{\partial}{\partial\theta}\alpha(\mathbf{r},t)|_{\theta=\pi,0} = 0\,. \tag{2}$$

In the rectangular computational domain that we use to solve the problem, the grid (r,θ) extends from $0\to R=$ 30a.u. in r (the density is negligible this far from the nucleus) and $0\to\pi$ in θ. Consequently the boundary conditions, Eq. (2) translate to

$$\begin{aligned} &\alpha(r=R,\theta,\varphi,t) = 0 \\ &\frac{\partial}{\partial\theta}\alpha(r,\theta,\varphi,t)|_{\theta=\pi} = 0 \qquad \frac{\partial}{\partial\theta}\alpha(r,\theta,\varphi,t)|_{\theta=0} = 0 \end{aligned} \tag{3}$$

The finite difference approximation of the derivative operator has the nice property that the resulting matrix is sparse, and consequently the operator, which by definition is local in space, remains so in this representation as well, since only a few adjacent grid points are coupled. The high sparsity of the matrix also allows for efficient computation of matrix inversion. Despite the computational efficiency it offers, caution is required to avoid numerical inaccuracies especially where the density becomes small. We ensure that our conclusions are robust with numerics, interpreting the results in regions where the inversion is accurate, and checking that the action of the matrix representing $\nabla\cdot n(\mathbf{r},t)\nabla$ on the solution vector $\alpha(\mathbf{r},t)$ agrees with the right-hand-side of Eq. (1) In a similar way, we calculate the exact Hartree potential $v_{\rm H}(\mathbf{r},t)$, by numerically inverting

$$\nabla^2 v_{\rm H}(\mathbf{r},t) = -4\pi n(\mathbf{r},t) \tag{4}$$

and then use $v_{\rm XC}(\mathbf{r},t) = v_{\rm S}(\mathbf{r},t) - v_{\rm H}(\mathbf{r},t) - v_{\rm ext}(\mathbf{r},t)$ to obtain the xc potential, and we isolate the correlation potential noting that, for our choice of KS state, $v_{\rm X}(\mathbf{r},t) = -v_{\rm H}(\mathbf{r},t)/2$.

II. MOVIES

As part of supplementary material, we provide the following two movies:

1. **DenVc.mp4** depicts the dynamics of correlation potential $v_{\rm C}(\mathbf{r},t)$ along $(\phi=0,\theta=\frac{\pi}{4})$ and $(\phi=0,\theta=\frac{3\pi}{4})$ in the lower left and right panels respectively, the corresponding density along those angles is displayed in the top panels.

2. **CurrentVec.mp4** shows the current density vector in the in the x-z half-plane.Note that the other half of the plane is symmetric to the one displayed in the movie.