

# Real-Time Approach to the Dynamical Bethe-Salpeter Equation for Finite Systems

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We present a real-time linear-response approach to solving the dynamical Bethe-Salpeter equation (BSE). The polarization part of the screened interaction is obtained from time-dependent Hartree propagation of the one-particle density matrix within an orbital basis-set representation. The frequency convolution defining the dynamical kernel is evaluated as a product in time, avoiding numerical integration and storage of the full screened Coulomb operator. The resulting nonlinear eigenvalue problem is then solved directly, going beyond the static screening approximation with full-frequency dependence in the screened interaction. While the deterministic scaling remains  $\mathcal{O}(N^6)$ , the time propagation formulation is readily compatible with grid-based stochastic sampling methods, which will open the possibility for dynamical BSE calculations of very large systems.

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## I. INTRODUCTION

Developing computationally efficient methods that accurately describe molecular excited states remains an active area of research. Coupled-cluster and multiconfigurational wavefunction methods can provide very accurate results for small molecules, but their computational scaling limits their applicability to large systems. Time-dependent density-functional theory (TDDFT) is considerably less expensive and is widely used for first-principles excited-state calculations. However, the standard adiabatic approximation neglects memory effects by assuming that the exchange-correlation kernel is local in time.<sup>1</sup> Consequently, standard adiabatic exchange-correlation kernels can struggle to describe excitation processes that depend explicitly on frequency, including states with substantial multiple-excitation character.<sup>2–8</sup>

Green’s-function-based many-body perturbation theory provides an alternative route to molecular excited states.<sup>9–12</sup> Within this framework, one begins from a mean-field ground-state description often obtained with density-functional or Hartree-Fock theory. Quasiparticle spectra, ionization potentials, and electron affinities can be calculated within the *GW* approximation, while neutral optical excitations are obtained by solving the two-particle Bethe-Salpeter equation (BSE). The BSE includes excitonic effects through an explicit electron-hole interaction kernel. The combined *GW*-BSE approach has become increasingly popular for studying molecular systems with its ability to describe valence, charge-transfer, and Rydberg excitations more consistently than TDDFT, even rivaling CCSD and CASPT2 methods.<sup>9,11–16</sup>

Most implementations of the BSE employ the static-screening approximation for the screened Coulomb interaction entering the electron-hole kernel. In this approximation, the frequency-dependent dynamical interaction  $\tilde{W}(\Omega)$  is replaced by the static interaction  $W(\omega = 0)$ . This assumes that the electron-hole attraction is instantaneous. We emphasize that this approximation applies specifically to the screened interaction entering the BSE kernel; the frequency dependence of  $W$  is generally retained in the preceding *GW* calculation of the quasiparticle self-energy. This approximation has enabled a large body of successful calculations for a wide variety of systems.<sup>10,12,13,17–20</sup> The static approximation is expected to be accurate when the screening response is much faster than the electron-hole dynamics. This translates to excitation energies that are well separated from the characteristic screen-

ing modes, i.e., plasmons, of the system. Dynamical effects can become important when this separation of energy scales breaks down, as demonstrated for copper and silver metals in Ref.<sup>21</sup>.

The static approximation nevertheless represents a physical restriction because it removes the delay, or memory, in the polarization response of the electronic system. This restriction can be especially problematic in finite systems.<sup>13,22</sup> Molecules generally exhibit weak screening and can have large exciton binding energies, such that the poles of  $W(\omega)$  correspond to molecular polarization modes that need not be energetically well separated from the optical excitation energies. In this regime, dynamical screening may not be adequately represented as a small correction to a static interaction. The static approximation can result in errors in energies on the order of 0.1 – 0.3 eV.<sup>23</sup> In general, the static *GW*-BSE performs best for spin-conserved transitions, with a poor description of triplet states.<sup>15,18</sup>

Several approaches have been developed to incorporate frequency dependence into the BSE. Perturbative dynamical corrections evaluate the frequency-dependent interaction using eigenstates obtained from a static BSE calculation.<sup>10,24</sup> Various plasmon-pole models have been developed for both solids and molecules.<sup>13,23,25–27</sup> An alternative formulation is to convert the frequency-dependence into a larger static eigenvalue problem.<sup>28</sup> These studies have demonstrated that dynamical screening tends to redshift singlet and triplet excitation energies and generally improves numerical agreement with high-level quantum chemical benchmarks.<sup>24,26,28</sup>

In principle, a frequency-dependent kernel can also introduce additional poles in the response function. This property provides a possible route to states with double- or higher-excitation character that are absent from the single-particle transition space of a static BSE or adiabatic TDDFT calculation.<sup>2,4,5,8,29,30</sup> Such effects are relevant in systems with open-shell ground-states as well as excited states with double-excitation character.<sup>4,31–33</sup>

Practical solutions of the nonlinear dynamical BSE remain computationally demanding because the screened interaction must be constructed and the BSE matrix diagonalized over many sampled frequencies.<sup>4–7,28,34,35</sup> In a conventional grid-based frequency-domain implementation, the independent-particle irreducible polarizability  $\chi_0(\omega)$  is evaluated through a sum-over-states expression and the random phase approximation (RPA) dielectric matrix is inverted to obtain  $W(\omega)$ . In general, the explicit evaluation and storage of the screened Coulomb operator over a dense frequency grid can become a major CPU and memory bot-

tleneck. We do note that various efficient methods that avoid dielectric matrix inversion for  $GW$ <sup>36–40</sup> and static  $GW$ -BSE<sup>17,41–43</sup> have been developed.

In this work, we present a real-time linear-response method for building the frequency-dependent interaction kernel entering the dynamical BSE for finite systems. The retarded reducible RPA response is obtained by propagating the time-dependent Hartree (TDH) equations following impulsive perturbations generated from orbital pair densities. The resulting induced density is contracted with the Coulomb tensor to obtain the required screened-interaction matrix elements. After constructing the corresponding time-ordered interaction, the convolution defining  $\tilde{W}(\Omega)$  is evaluated as a product in the time domain followed by a Fourier transform. This avoids explicit numerical integration over frequency and does not require storage of the full real-space screened Coulomb operator. We validate the real-time formalism in two ways. First, we verify that the static BSE obtained from the real-time TDH response reproduces an independent implementation based on static dielectric-matrix inversion. Second, we verify that the time-product evaluation of  $\tilde{W}(\Omega)$  reproduces direct numerical evaluation of the frequency convolution using the same  $W(\omega)$  obtained from the real-time response function.

The present formulation is the dynamical extension of our earlier stochastic real-time approaches for representing the screened Coulomb matrix in large molecular systems.<sup>38,41,42,44–47</sup> Although the deterministic orbital-basis implementation employed here retains the formal  $\mathcal{O}(N^6)$  scaling, the real-time construction is directly compatible with stochastic sampling and grid-based propagation.

The paper is organized as follows. We first present the theoretical formulation and real-time implementation, followed by validation for the silane molecule,  $\text{SiH}_4$ . We conclude with a discussion of future directions and the extension of the real-time approach to much larger systems.

## II. THEORY

In this section, we develop the real-time method for evaluating the dynamically screened Coulomb interaction,  $\tilde{W}(\Omega)$ . Both the software implementation and results presented here employ the Tamm-Dancoff Approximation (TDA) to the excitonic BSE Hamiltonian. The resonant-antiresonant coupling elements of  $\tilde{W}$  can produce numerical divergences<sup>24</sup> and will

be explored in future work.

## A. Dynamical BSE

The BSE is cast as a nonlinear eigenvalue problem in the space of  $N_v N_c$  valence-to-conduction transitions:

$$\tilde{A}_{ia,jb}(\Omega) = (\varepsilon_a - \varepsilon_i + \Delta)\delta_{ij}\delta_{ab} + \kappa(ia|jb) - \tilde{W}_{ab,ij}(\Omega), \quad (1)$$

where  $\varepsilon_i$  are occupied (valence) and  $\varepsilon_a$  unoccupied (conduction) orbital energies. In principle, these energies come from  $GW$  theory. Here, we apply a rigid scissor  $\Delta$  to local density approximation (LDA)-DFT<sup>48</sup> eigenvalue differences to match the quasiparticle gap for SiH<sub>4</sub> of Louie and Rohlifing reported in Ref.<sup>13</sup>. The reference  $GW$  gap uses LDA-DFT functions and a first-order perturbative correction for the self-energy. The second term is the repulsive exciton exchange interaction responsible for singlet-triplet splitting, with  $\kappa = 0, 2$  for triplet and singlet excitations, respectively. Two-electron Coulomb integrals  $(pq|rs)$  are expressed in  $(rr|r'r')$  notation:

$$(pq|rs) = \int dr' dr \phi_p(r) \phi_q(r) |r - r'|^{-1} \phi_r(r') \phi_s(r'). \quad (2)$$

Throughout the manuscript, occupied molecular orbitals (MOs) are labeled  $i, j, k, l$ , virtual MOs by  $a, b, c, d$ , and general MOs by  $p, q, r, s$ . The direct electron-hole interaction has matrix elements:

$$\tilde{W}_{ab,ij}(\Omega) = \frac{i}{2\pi} \int d\omega W_{ab,ij}(\omega) \times \left[ \frac{1}{\Omega - (\varepsilon_b - \varepsilon_i + \Delta + \omega) + i\eta} + \frac{1}{\Omega - (\varepsilon_a - \varepsilon_j + \Delta - \omega) + i\eta} \right], \quad (3)$$

where  $W_{ab,ij}(\omega) = \int dr' dr \phi_a(r) \phi_b(r) W(r, r', \omega) \phi_i(r') \phi_j(r')$ , and  $\eta$  is the numerical broadening, taken here to be  $\approx 50$  meV. Eq. (3) is a Shindo-like approximation, where the frequency dependence of the BSE is factorized separately into the screened interaction and non-interacting electron-hole propagators.<sup>4,11,22,25</sup> A derivation of Eq. (1) is provided in Appendix A. While calculating Eq. (1) is relatively straightforward, it is numerically very expensive to build the real-space  $W(r, r', \omega)$  operator over many frequencies and then perform the  $\mathcal{O}(N_\Omega N_\omega)$  numerical integration over  $\mathcal{O}(N_v^2 N_c^2)$  electron-hole matrix elements in Eq. (3).

Solution of the nonlinear eigenvalue problem:

$$\tilde{A}(\Omega)X_n(\Omega) = \lambda_n(\Omega)X_n(\Omega), \quad (4)$$

yields physical excitation energies that satisfy  $\text{Re } \lambda_n(\Omega) = \Omega$ . We obtain the roots of Eq. (4) by a secant interpolation method, which we discuss later.

The standard static BSE is obtained by replacing  $\tilde{W}_{ab,ij}(\Omega) \approx W_{ab,ij}(\omega = 0)$ , reducing the nonlinear eigenproblem to a linear one. In this work, we present results for both the static and dynamic BSE.

## B. Real-Time Based Approach to $\tilde{W}_{ab,ij}(\Omega)$

We outline how the electron-hole kernel is constructed through time-propagation. Rather than explicitly performing the frequency convolution in Eq. (3), we calculate the polarization part of the screened Coulomb interaction through real-time linear response theory and evaluate the frequency convolution as a product in the time domain.

It is useful to first separate the time-ordered screened Coulomb interaction into its instantaneous and polarization parts,

$$W_{ab,ij}(t) = (ab|ij)\delta(t) + W_{ab,ij}^{\text{pol}}(t). \quad (5)$$

The real-time propagation is used to obtain the reducible RPA polarization function<sup>38,49</sup>, from which the polarization part of the screened interaction is constructed, while the instantaneous bare Coulomb term is added separately.

The real-time approach begins with a finite orbital basis. We expand the time-dependent orbitals as

$$\psi_p(r, t) = \sum_s C_{sp}(t)\phi_s(r), \quad (6)$$

where the coefficient matrix contains both occupied and unoccupied orbitals. For simplicity, we consider a single perturbing occupied-orbital pair  $kl$ , with the TDA BSE containing a total of  $N_v(N_v + 1)/2$  unique pairs. The corresponding perturbing potential produced by  $kl$  is represented in the orbital basis by

$$D_{pq}^{kl} = (pq|kl), \quad (7)$$

and an impulsive perturbation of strength  $\lambda$  is applied:

$$C^{kl}(t = 0^+) = e^{-i\lambda D^{kl}} C(t = 0), \quad (8)$$

where  $\lambda$  is taken to be 0.0001. Here, we diagonalize each  $D^{kl}$ ; however, one could also use a normalized first-order Taylor expansion, i.e.,  $C^{kl}(t = 0^+) \approx \mathcal{N}(1 - i\lambda D^{kl})C(t = 0)$ , where  $\mathcal{N}$  is a normalization factor with a correction quadratic in  $\lambda$ .

The perturbed coefficient matrix is propagated according to

$$i \frac{\partial C^{kl}(t)}{\partial t} = H^{kl}(t) C^{kl}(t), \quad (9)$$

where the density matrix is

$$P_{pq}^{kl}(t) = \sum_m C_{pm}^{kl}(t) f_m C_{qm}^{kl,*}(t), \quad (10)$$

where  $f_m$  denotes the orbital occupation. To obtain the screened Coulomb interaction at the RPA level, we use the time-dependent Hartree Hamiltonian,

$$H_{pq}^{kl}(t) = \varepsilon_p \delta_{pq} + 2 \sum_{rs} (pq|rs) [P_{rs}^{kl}(t) - P_{rs}(t = 0)], \quad (11)$$

where the factor of two accounts for spin. The induced density matrix associated with the perturbing source  $kl$  is

$$\delta P_{pq}^{kl}(t) = \frac{1}{\lambda} [P_{pq}^{kl}(t) - P_{pq}(t = 0)]. \quad (12)$$

The resulting retarded polarization contribution to the screened interaction is

$$W_{pq}^{\text{pol},kl,R}(t) = 2 \sum_{rs} (pq|rs) \delta P_{rs}^{kl}(t). \quad (13)$$

The causal frequency-domain response is obtained by applying a Gaussian damping and Fourier transforming the positive-time response,

$$W_{pq}^{\text{pol},kl,R}(\omega) = \int_0^\infty dt e^{-\gamma^2 t^2 / 2} e^{i\omega t} W_{pq}^{\text{pol},kl,R}(t). \quad (14)$$

For the calculations reported here, the positive-time response is propagated to  $T_{\text{max}} \approx 20$  fs, corresponding to a Gaussian damping parameter  $\gamma = 3/T_{\text{max}} \approx 100$  meV. A time-step of  $dt = 0.05$  au is used for all simulations.

For real-valued perturbations, the time-ordered and retarded interactions are related in frequency space by<sup>50</sup>

$$W_{pq}^{\text{pol},kl}(\omega) = \begin{cases} W_{pq}^{\text{pol},kl,R}(\omega), & \omega \geq 0, \\ [W_{pq}^{\text{pol},kl,R}(\omega)]^*, & \omega < 0. \end{cases} \quad (15)$$

The time-ordered interaction on the full time axis is then obtained from

$$W_{pq}^{\text{pol},kl}(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega e^{-i\omega t} W_{pq}^{\text{pol},kl}(\omega). \quad (16)$$

This causal-to-time-ordered transformation follows the real-time prescription used previously in stochastic *GW* calculations.<sup>38,44</sup>

For a TDA BSE matrix element  $\tilde{W}_{ab,ij}(\Omega)$ , the occupied-occupied perturbing source is  $kl = ij$  and the response is measured in the unoccupied-unoccupied sector  $pq = ab$ . The polarization contribution to the dynamical interaction can then be evaluated directly in the time domain as

$$\begin{aligned} \tilde{W}_{ab,ij}^{\text{pol}}(\Omega) = & \int_0^{\infty} dt W_{ab}^{\text{pol},ij}(t) e^{-i(\varepsilon_b - \varepsilon_i + \Delta)t} e^{i\Omega t} e^{-\eta|t|} \\ & + \int_{-\infty}^0 dt W_{ab}^{\text{pol},ij}(t) e^{i(\varepsilon_a - \varepsilon_j + \Delta)t} e^{-i\Omega t} e^{-\eta|t|}. \end{aligned} \quad (17)$$

Eq. (17) is the central real-time expression used in our implementation. The orbital-energy phase factors originate from the energy denominators in Eq. (3). The frequency convolution over  $\omega$  is therefore replaced by multiplication in the time domain followed by Fourier transformation.

The full dynamically screened interaction entering the BSE is obtained by adding the instantaneous bare Coulomb contribution,

$$\tilde{W}_{ab,ij}(\Omega) = (ab|ij) + \tilde{W}_{ab,ij}^{\text{pol}}(\Omega). \quad (18)$$

The same real-time propagation also gives the zero-frequency screened interaction used for the static BSE,

$$W_{ab,ij}(\omega = 0) = (ab|ij) + \int_0^{\infty} dt e^{-\gamma^2 t^2 / 2} W_{ab}^{\text{pol},ij,R}(t). \quad (19)$$

The above procedure is repeated for each unique occupied-orbital pair. This task is trivially parallelizable over separate MPI processes, so time propagation for different perturbations can be performed simultaneously.<sup>38,42,44,45</sup>

### III. VALIDATION OF THE METHOD

We validate the real-time formalism for the silane molecule,  $\text{SiH}_4$ , in two steps. First, we verify that the static BSE obtained from the real-time TDH response reproduces an independent implementation based on static dielectric matrix inversion. Second, we verify that

the time-product evaluation of  $\tilde{W}(\Omega)$  reproduces the numerical evaluation of the frequency convolution in Eq. (3). We then compare our results qualitatively with those presented by Rohlfing and Louie in Ref.<sup>13</sup>.

We perform plane-wave LDA-DFT calculations on a finite grid of  $N_{\text{grid}} = 16^3 = 4096$  points with an isotropic grid spacing of  $dx = dy = dz = 0.6$  Bohr.<sup>48</sup> We utilize Troullier-Martins norm-conserving pseudopotentials<sup>51</sup> to account for the effect of the core electrons on the electron-nucleus attraction. The Martyna-Tuckerman approach for electron-electron interactions is used to reduce periodic images.<sup>52</sup> The LDA-DFT calculation gives a HOMO-LUMO gap of 6.61 eV. We then apply a scissor correction of  $\Delta = 7.18$  eV to the bare transition energies entering the BSE Hamiltonian, reproducing the diagonal *GW* gap of 13.79 eV reported in Ref.<sup>13</sup>.

We plot in Fig. 1 the static BSE spectrum obtained from the real-time method against that obtained using a standard frequency-domain construction of  $W_{ab,ij}(\omega = 0)$ . Both calculations use all  $N_v = 4$  occupied valence orbitals and  $N_c = 24$  unoccupied conduction orbitals. We obtain excellent agreement between the two implementations, with mean absolute deviations below 1 meV across the static BSE eigenvalue spectrum.

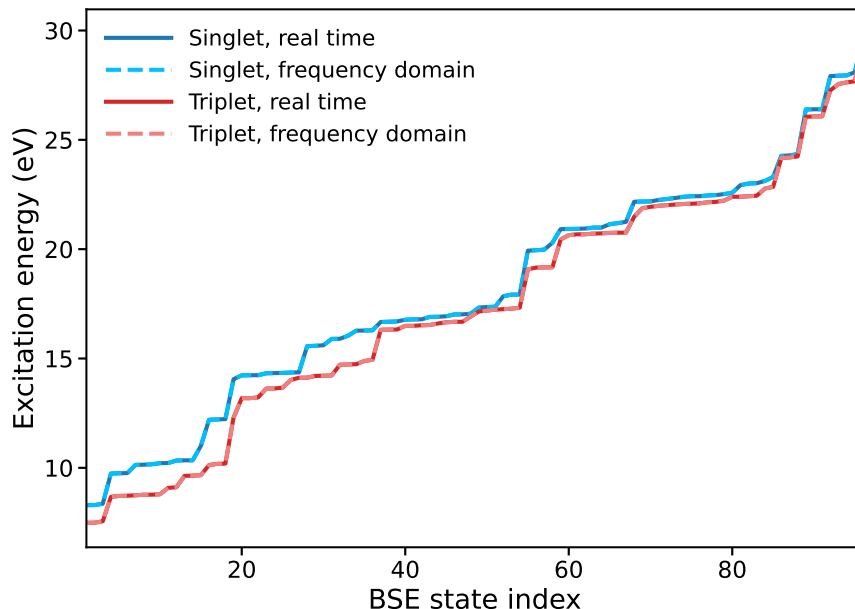


FIG. 1. Static BSE excitation energies of  $\text{SiH}_4$  obtained from real-time TDH response (Eq. (19)) vs. inverting the static RPA dielectric matrix. The excitation energies are plotted as a function of the state index for the singlet and triplet manifolds.

We find the dynamical roots from the eigenvalues obtained at each sampled frequency  $\Omega$  using secant interpolation.<sup>25</sup> We define  $F_n(\Omega) = \text{Re } \lambda_n(\Omega) - \Omega$ , then a root is identified when  $F_n(\Omega_j)F_n(\Omega_{j+1}) < 0$  between two neighboring frequencies. The excitation energy is then estimated as  $\Omega_n \approx \Omega_j - F_n(\Omega_j) \frac{\Omega_{j+1} - \Omega_j}{F_n(\Omega_{j+1}) - F_n(\Omega_j)}$ . The resulting root satisfies  $\text{Re } \lambda_n(\Omega_n) = \Omega_n$ .

|              | <b>Static</b>       | <b>Time Product</b> |                       | <b>Frequency Convolution</b> |                       | <b>Ref.<sup>13</sup></b> |         |                       |
|--------------|---------------------|---------------------|-----------------------|------------------------------|-----------------------|--------------------------|---------|-----------------------|
| <b>State</b> | $E_{\text{static}}$ | $E_{\text{dyn}}$    | $\Delta_{\text{dyn}}$ | $E_{\text{dyn}}$             | $\Delta_{\text{dyn}}$ | Static                   | Dynamic | $\Delta_{\text{dyn}}$ |
| Singlet      | 8.29                | 8.17                | -0.12                 | 8.17                         | -0.12                 | 9.24                     | 9.16    | -0.08                 |
| Triplet      | 7.48                | 7.14                | -0.35                 | 7.14                         | -0.35                 | 8.64                     | 8.51    | -0.13                 |

TABLE I. Lowest singlet and triplet excitation energies of SiH<sub>4</sub> obtained from the static and dynamical BSE using the Time Product vs. Frequency Convolution methods for evaluating  $\tilde{W}(\Omega)$ . All energies are in eV. The dynamical correction is defined as  $\Delta_{\text{dyn}} = E_{\text{dyn}} - E_{\text{static}}$ . Results from Ref.<sup>13</sup> are included for comparison. In Ref.<sup>13</sup>, dynamical screening is treated using an iterative plasmon-pole representation of  $\tilde{W}(\Omega)$  and the BSE matrix is constructed from *GW* quasiparticle states, whereas the present calculations use LDA states and evaluate the full-frequency screened interaction.

We next validate the time-product transformation introduced in Eq. (17). Here, the dynamical interaction is evaluated in two mathematically equivalent ways. In the Time Product approach, the frequency convolution is replaced by multiplication in the time domain followed by Fourier transformation according to Eq. (17). In the Frequency Convolution approach, the same dynamical interaction is instead obtained by directly performing the numerical frequency integral in Eq. (3). This comparison isolates the numerical transformation from  $W(\omega)$  to  $\tilde{W}(\Omega)$  and tests whether the time-product formulation reproduces the frequency convolution.

In Table I, we report the lowest-lying singlet and triplet excitation energies of SiH<sub>4</sub> using the BSE with both static and dynamical screening of the electron-hole interaction. We also include the results of Rohlfing and Louie from Ref.<sup>13</sup>. In both calculations, dynamical screening produces a redshift of the lowest excitation energies. The reference values are not directly comparable because Ref.<sup>13</sup> utilizes quasiparticle *GW* states rather than LDA-DFT Kohn-Sham states in the BSE calculation. The reference comparison is used to establish the expected direction and approximate magnitude of the dynamical correction. The larger

redshift of the triplet excitation energy in the present calculation reflects the absence of the repulsive exchange contribution for the triplet, such that changes to the direct screened interaction have a larger effect on the excitation energy. Further, in Ref.<sup>13</sup> it is discussed that the use of quasiparticle *GW* states, rather than LDA-DFT ones, weakens the strength of the direct electron-hole interaction, resulting in higher absolute energies.

#### IV. CONCLUSIONS AND FUTURE DIRECTIONS

We have formulated a real-time approach for constructing the frequency-dependent screened interaction entering the dynamical BSE. A pair-density source perturbation is used to generate the retarded screened interaction through TDH propagation. The causal response is transformed to a time-ordered interaction, multiplied by phase factors representing non-interacting electron-hole pair propagation, and Fourier transformed to obtain  $\tilde{W}(\Omega)$ . The resulting nonlinear TDA BSE is solved by diagonalizing the excitonic matrix on a grid of trial frequencies and locating roots numerically. For  $\text{SiH}_4$ , we find that the matrix  $\tilde{A}(\Omega)$  varies smoothly with  $\Omega$ , implying that interpolation approaches work well and one can use only a small number of sampling frequencies  $\Omega$ .

It is challenging to include the off-diagonal block of the dynamical BSE because of numerical divergences.<sup>24</sup> We will explore how to handle this issue in future work. It is also worth mentioning that utilizing a *GW* approximation to the single-particle self-energy can result in unphysical solutions to the BSE. This has been explored by others<sup>4-6,49,53</sup>, and we will consider this in future work. Here, we targeted numerical validation of the real-time formulation and the effect of dynamical screening on the lowest-lying excitations of a small molecule.

This method maintains the formal  $\mathcal{O}(N^6)$  scaling, but the real-time formulation opens two related future directions: applications to much larger systems through stochastic methods and a possible recasting as a memory-based time evolution. We note that related real-time density-matrix approaches have incorporated an RPA-screened Coulomb interaction to evaluate electron-electron scattering rates within a quantum Boltzmann equation.<sup>54</sup>

We briefly discuss the large-scale stochastic extension below, while the memory-based formulation is outlined in Appendix B.

## A. Future Applications with Stochastic Methods

The real-time formulation lends itself naturally to the stochastic methods we have developed in Refs.<sup>38,41,42,44–46,55–57</sup>. Rather than explicitly constructing all matrix elements of the frequency-dependent screened interaction in the orbital basis, only a small set of stochastic matrix elements could be calculated by using random linear combinations of the occupied and unoccupied orbitals. The corresponding polarization response can then be measured using stochastic real-time dynamics.<sup>38,58</sup>

We illustrate briefly how the dynamical BSE could be brought to systems containing hundreds to potentially thousands of valence electrons. We have shown with Eq. (17) that the matrix elements of  $\tilde{W}(t)$  and  $W(t)$  are related by

$$\tilde{W}_{ab,ij}(t) = W_{ab,ij}(t)e^{-\eta|t|} \left[ e^{-i(\varepsilon_b - \varepsilon_i + \Delta)t} \theta(t) + e^{i(\varepsilon_a - \varepsilon_j + \Delta)t} \theta(-t) \right]. \quad (20)$$

This relation is particularly useful for stochastic sampling because the orbital-energy phase factors can be transferred to random orbitals restricted to the occupied and unoccupied subspaces. The corresponding sampled matrix element can be written as

$$\begin{aligned} (\bar{\gamma}\bar{\gamma}|\tilde{W}(t)|\bar{\beta}\bar{\beta}) &= \theta(t)e^{-i\Delta t}e^{-\eta|t|}(\bar{\gamma}\bar{\gamma}(t)|W(t)|\bar{\beta}(-t)\bar{\beta}) \\ &+ \theta(-t)e^{i\Delta t}e^{-\eta|t|}(\bar{\gamma}(-t)\bar{\gamma}|W(t)|\bar{\beta}\bar{\beta}(t)). \end{aligned} \quad (21)$$

Here,  $\bar{\gamma}$  and  $\bar{\gamma}$  denote random-sign linear combinations of MOs within the unoccupied subspace, while  $\bar{\beta}$  and  $\bar{\beta}$  denote random linear combinations of MOs within the occupied subspace. For example,  $\bar{\gamma}(r) = \sum_{a \in N_c} \pm \phi_a(r)$ , with an analogous expression for a random occupied function. The time dependence of these random states is governed by the corresponding one-particle Hamiltonian, i.e.,  $|\bar{\gamma}(t)\rangle = e^{-iH_0 t}|\bar{\gamma}(0)\rangle$ , where  $H_0$  may be an accurate quasiparticle Hamiltonian obtained from an optimally tuned range-separated hybrid DFT reference<sup>57</sup>. The scissor shift  $\Delta$  present in Eqs. (20)-(21) can be calculated with the low-scaling stochastic *GW* method.<sup>59,60</sup> The resulting time-dependent matrix elements can then be Fourier transformed to obtain the corresponding sampled matrix elements of  $\tilde{W}(\Omega)$ .

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## DECLARATION OF COMPETING INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

## Appendix A: Derivation of the Dynamical Bethe-Salpeter Equation within the Tamm-Dancoff and Shindo Approximations

We begin from the Schwinger functional-derivative technique for the two-particle correlation function.<sup>11,22</sup> First, we introduce an external potential  $U(1, 2)$ , and define

$$L(1, 2; 1', 2') = -i \frac{\delta G(1, 1')}{\delta U(2', 2)} \quad (\text{A1})$$

The inverse Green's function is then:

$$G^{-1}(1, 2) = G_0^{-1}(1, 2) - U(1, 2) - v_H(1)\delta(1, 2) - \Sigma(1, 2), \quad (\text{A2})$$

where  $v_H(1) = -i \int d2 v(1, 2)G(2, 2^+)$  is the Hartree potential and  $\Sigma$  is the single-particle self-energy, encompassing all many-body exchange-correlation effects. By using the identity  $\frac{\delta}{\delta U}(GG^{-1}) = 0$  and taking the external potential to zero, one arrives at the Bethe-Salpeter equation

$$\begin{aligned} L(1, 2; 1', 2') &= L_0(1, 2; 1', 2') \\ &+ \int d3 d4 d5 d6 L_0(1, 4; 1', 3)\Xi(3, 5; 4, 6)L(6, 2; 5, 2'), \end{aligned} \quad (\text{A3})$$

where

$$L_0(1, 2; 1', 2') = -iG(1, 2')G(2, 1') \quad (\text{A4})$$

and the BSE kernel is defined as:

$$\Xi(3, 5; 4, 6) = i \frac{\delta [v_H(3)\delta(3, 4) + \Sigma(3, 4)]}{\delta G(6, 5)}. \quad (\text{A5})$$

Within the  $GW$  approximation,

$$\Sigma(3, 4) = iG(3, 4)W(3^+, 4). \quad (\text{A6})$$

Neglecting the functional derivative  $\delta W/\delta G$  produces

$$\begin{aligned} \Xi(3, 5; 4, 6) &= \delta(3, 4)\delta(5, 6)v(3, 6) \\ &\quad - \delta(3, 6)\delta(4, 5)W(3, 4). \end{aligned} \quad (\text{A7})$$

The first term produces the repulsive bare exchange interaction, while the second produces the attractive screened direct interaction.

Following the frequency convention of Romaniello *et al.*,<sup>4</sup> the equilibrium four-point function is written in terms of an external bosonic frequency  $\Omega$  and two internal fermionic frequencies  $\omega'$  and  $\omega''$ . Suppressing spatial and spin variables, the BSE becomes

$$\begin{aligned} L(\Omega, \omega', \omega'') &= L_0(\Omega, \omega', \omega'') \\ &\quad - iG\left(\omega' + \frac{\Omega}{2}\right) G\left(\omega' - \frac{\Omega}{2}\right) v \int \frac{d\bar{\omega}}{2\pi} L(\Omega, \bar{\omega}, \omega'') \\ &\quad + iG\left(\omega' + \frac{\Omega}{2}\right) G\left(\omega' - \frac{\Omega}{2}\right) \int \frac{d\bar{\omega}}{2\pi} W(\omega' - \bar{\omega}) L(\Omega, \bar{\omega}, \omega''), \end{aligned} \quad (\text{A8})$$

with

$$\begin{aligned} L_0(\Omega, \omega', \omega'') &= -2\pi i \delta(\omega' - \omega'') \\ &\quad \times G\left(\omega' + \frac{\Omega}{2}\right) G\left(\omega'' - \frac{\Omega}{2}\right). \end{aligned} \quad (\text{A9})$$

We now project onto the resonant electron-hole sector and assume real-valued orbitals. The projected correlation function is then:

$$\begin{aligned} L_{ia,jb}(\Omega, \omega', \omega'') &= \int dx_1 dx_2 dx_{1'} dx_{2'} \phi_i(x_1) \phi_a(x_{1'}) \\ &\quad \times L(x_1, x_2; x_{1'}, x_{2'}; \Omega, \omega', \omega'') \phi_j(x_{2'}) \phi_b(x_2). \end{aligned} \quad (\text{A10})$$

The Tamm-Dancoff BSE in this basis is

$$\begin{aligned} L_{ia,jb}(\Omega, \omega', \omega'') &= L_{ia,jb}^0(\Omega, \omega', \omega'') - iG_a\left(\omega' + \frac{\Omega}{2}\right) G_i\left(\omega' - \frac{\Omega}{2}\right) \\ &\quad \times \sum_{kc} \int \frac{d\omega_1}{2\pi} [\kappa(ia|kc) - (ac|W(\omega' - \omega_1)|ik)] L_{kc,jb}(\Omega, \omega_1, \omega''), \end{aligned} \quad (\text{A11})$$

where  $\kappa = 2$  for singlet excitations and  $\kappa = 0$  for triplet excitations. The uncorrelated propagator is

$$L_{ia,jb}^0(\Omega, \omega', \omega'') = -2\pi i \delta(\omega' - \omega'') \delta_{ij} \delta_{ab} \times G_a\left(\omega' + \frac{\Omega}{2}\right) G_i\left(\omega'' - \frac{\Omega}{2}\right). \quad (\text{A12})$$

The quasiparticle Green's functions are

$$G_a(z) = \frac{1}{z - \varepsilon_a + i\eta}, \quad G_i(z) = \frac{1}{z - \varepsilon_i - i\eta}, \quad (\text{A13})$$

where  $\eta \rightarrow 0^+$ . We define the correlation function with the internal fermionic frequencies integrated out,

$$\tilde{L}_{ia,jb}(\Omega) = \int \frac{d\omega' d\omega''}{(2\pi)^2} L_{ia,jb}(\Omega, \omega', \omega''). \quad (\text{A14})$$

With frequency dependence in  $W$ , this integration does not close the BSE. We therefore adopt the Shindo-type approximation<sup>11,22,25</sup>

$$L_{ia,jb}(\Omega, \omega', \omega'') \approx \frac{G_a(\omega' + \frac{\Omega}{2}) G_i(\omega' - \frac{\Omega}{2})}{N_{ai}(\Omega)} \tilde{L}_{ia,jb}(\Omega) \times \frac{G_b(\omega'' + \frac{\Omega}{2}) G_j(\omega'' - \frac{\Omega}{2})}{N_{bj}(\Omega)}, \quad (\text{A15})$$

where

$$N_{ai}(\Omega) = \int \frac{d\bar{\omega}}{2\pi} G_a\left(\bar{\omega} + \frac{\Omega}{2}\right) G_i\left(\bar{\omega} - \frac{\Omega}{2}\right) = \frac{i}{\Omega - (\varepsilon_a - \varepsilon_i) + i\eta}. \quad (\text{A16})$$

The same expression, with  $a, i$  replaced by  $b, j$ , defines  $N_{bj}(\Omega)$ . The normalization ensures that integration of Eq. (A15) over  $\omega'$  and  $\omega''$  recovers  $\tilde{L}_{ia,jb}(\Omega)$ .

The non-interacting contribution is

$$\tilde{L}_{ia,jb}^0(\Omega) = \frac{\delta_{ij} \delta_{ab}}{\Omega - (\varepsilon_a - \varepsilon_i) + i\eta}, \quad (\text{A17})$$

while the bare exchange part is

$$\tilde{L}_{ia,jb}^x(\Omega) = \frac{1}{\Omega - (\varepsilon_a - \varepsilon_i) + i\eta} \sum_{kc} \kappa(ia|kc) \tilde{L}_{kc,jb}(\Omega). \quad (\text{A18})$$

For the screened direct term, inserting the Shindo ansatz gives

$$\begin{aligned}\tilde{L}_{ia,jb}^d(\Omega) &= i \sum_{kc} \frac{\tilde{L}_{kc,jb}(\Omega)}{N_{ck}(\Omega)} \int \frac{d\omega' d\omega_1}{(2\pi)^2} \\ &\quad \times G_a\left(\omega' + \frac{\Omega}{2}\right) G_i\left(\omega' - \frac{\Omega}{2}\right) (ac|W(\omega' - \omega_1)|ik) \\ &\quad \times G_c\left(\omega_1 + \frac{\Omega}{2}\right) G_k\left(\omega_1 - \frac{\Omega}{2}\right).\end{aligned}\tag{A19}$$

Evaluating the two internal-frequency integrals by contour integration yields

$$\tilde{L}_{ia,jb}^d(\Omega) = -\frac{1}{\Omega - (\varepsilon_a - \varepsilon_i) + i\eta} \sum_{kc} \tilde{W}_{ia,kc}(\Omega) \tilde{L}_{kc,jb}(\Omega),\tag{A20}$$

where

$$\begin{aligned}\tilde{W}_{ia,kc}(\Omega) &\equiv \tilde{W}_{ac,ik}(\Omega) \\ &= \frac{i}{2\pi} \int_{-\infty}^{\infty} d\omega (ac|W(\omega)|ik) \\ &\quad \times \left[ \frac{1}{\Omega - (\varepsilon_c - \varepsilon_i + \omega) + i\eta} + \frac{1}{\Omega - (\varepsilon_a - \varepsilon_k - \omega) + i\eta} \right].\end{aligned}\tag{A21}$$

Equivalently, for the matrix element coupling the electron-hole pairs  $ia$  and  $jb$ ,

$$\begin{aligned}\tilde{W}_{ia,jb}(\Omega) &\equiv \tilde{W}_{ab,ij}(\Omega) \\ &= \frac{i}{2\pi} \int_{-\infty}^{\infty} d\omega W_{ab,ij}(\omega) \\ &\quad \times \left[ \frac{1}{\Omega - (\varepsilon_b - \varepsilon_i + \omega) + i\eta} + \frac{1}{\Omega - (\varepsilon_a - \varepsilon_j - \omega) + i\eta} \right].\end{aligned}\tag{A22}$$

Combining the uncorrelated, bare exchange, and screened direct terms gives

$$\begin{aligned}\tilde{L}_{ia,jb}(\Omega) &= \frac{\delta_{ij}\delta_{ab}}{\Omega - (\varepsilon_a - \varepsilon_i) + i\eta} \\ &\quad + \frac{1}{\Omega - (\varepsilon_a - \varepsilon_i) + i\eta} \sum_{kc} \left[ \kappa(ia|kc) - \tilde{W}_{ia,kc}(\Omega) \right] \tilde{L}_{kc,jb}(\Omega).\end{aligned}\tag{A23}$$

We define the frequency-dependent TDA BSE matrix as:

$$\tilde{A}_{ia,jb}(\Omega) = (\varepsilon_a - \varepsilon_i)\delta_{ij}\delta_{ab} + \kappa(ia|jb) - \tilde{W}_{ia,jb}(\Omega).\tag{A24}$$

Equation (A23) can then be written in matrix form:

$$\tilde{L}(\Omega) = \left[ (\Omega + i\eta)I - \tilde{A}(\Omega) \right]^{-1}.\tag{A25}$$

## Appendix B: Possible Memory-Based Time Evolution

An advantage of the time-based approach is that it can be used to physically evolve density matrices  $\rho(t)$ . Schematically, a “disturbed” density matrix at  $t = 0$  (for example, due to a dipole excitation),  $\rho(t = 0)$ , would be propagated in time as

$$\rho(t + dt) = U(t + dt, t)\rho(t)U^\dagger(t, t + dt) \quad (\text{B1})$$

where

$$U(t + dt, t) = \exp(-i\tilde{\mathbf{L}}(t)dt) \quad (\text{B2})$$

and  $\tilde{\mathbf{L}}(t)$  is a matrix projection of either the Tamm-Dancoff operator or the full BSE matrix including resonant and anti-resonant components, using the time-ordered  $W(t)$  components.  $\tilde{\mathbf{L}}(t)$  is based on  $\tilde{W}(t)$  from Eq. (20).

The propagated density matrix would be followed in time and then used to extract, e.g., the dipole response and absorption spectra using standard methods, or steady states for continuously disturbed systems.

This schematically outlined approach would be developed and investigated in future publications.

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