Time-Dependent Linear Response for Coupled Electron-Boson Systems

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Time-dependent linear response theory[1] is the most widely used approach for calculating excitation energies of electronic systems. In doing so, the Casida equation[2] is solved. This formalism is not restricted to electronic systems, but can also be applied to systems coupled to other fermionic particles (e.g. protons[3]), or to bosonic (quasi-)particles, such as photons[4], Drude oscillators, or nuclear vibrations:

$$\begin{pmatrix} \boldsymbol{A}^{\mathrm{I}} & \boldsymbol{A}^{\mathrm{I}\leftrightarrow\mathrm{II}} & \boldsymbol{B}^{\mathrm{I}} & \boldsymbol{B}^{\mathrm{I}\leftrightarrow\mathrm{II}} \\ (\boldsymbol{A}^{\mathrm{I}\leftrightarrow\mathrm{II}})^{\dagger} & \boldsymbol{A}^{\mathrm{II}} & (\boldsymbol{B}^{\mathrm{I}\leftrightarrow\mathrm{II}})^{\dagger} & \boldsymbol{B}^{\mathrm{II}} \\ -(\boldsymbol{B}^{\mathrm{I}})^{*} & -(\boldsymbol{B}^{\mathrm{I}\leftrightarrow\mathrm{II}})^{*} & -(\boldsymbol{A}^{\mathrm{I}})^{*} & -(\boldsymbol{A}^{\mathrm{I}\leftrightarrow\mathrm{II}})^{*} \\ -(\boldsymbol{B}^{\mathrm{I}\leftrightarrow\mathrm{II}})^{\mathrm{T}} & -(\boldsymbol{B}^{B})^{*} & -(\boldsymbol{A}^{\mathrm{I}\leftrightarrow\mathrm{II}})^{\mathrm{T}} & (-\boldsymbol{A}^{\mathrm{II}})^{*} \end{pmatrix} \begin{pmatrix} \boldsymbol{x}^{\mathrm{I}} \\ \boldsymbol{x}^{\mathrm{II}} \\ \boldsymbol{y}^{\mathrm{I}} \\ \boldsymbol{y}^{\mathrm{II}} \end{pmatrix} = \hbar\omega \begin{pmatrix} \boldsymbol{x}^{\mathrm{I}} \\ \boldsymbol{x}^{\mathrm{II}} \\ \boldsymbol{y}^{\mathrm{I}} \\ \boldsymbol{y}^{\mathrm{II}} \end{pmatrix} \tag{1}$$

Hereby, the matrices and vectors are defined in a basis of single-particle excitations. The $\boldsymbol{A}$ matrices and $\boldsymbol{x}$ vectors contain information about occupied-virtual excitations, while the $\boldsymbol{B}$ matrices and $\boldsymbol{y}$ vectors contain information about virtual-occupied excitations (i.e. deexcitations). The superscript denotes whether a submatrix/subvector contains contributions from subsystem I or II or whether it is an interaction term ($\mathrm{I} \leftrightarrow \mathrm{II}$). An extension to additional subsystems is straightforward.
Starting from a mean-field treatment, the A and B matrices consist of the occupied-virtual elements of the derivative of the Fock matrix w.r.t. the density matrix. With this methodology, e.g. environment effects can be accurately captured at a low computational cost.

On the poster, theoretical aspects, details of a general implementation and sample applications using the Hartree–Fock approximation for the electrons will be shown.

References

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