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Licensing

`SHARC`–v2.1 is licensed under the GNU general public license–v3. This license is reproduced in the Appendix A of this manual. The license requires that if we distribute copies of `SHARC`–v2.1, we must pass on to recipients the same freedoms that we received under the GNU published license, and we hereby do so.

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Acknowledgments

We requested permission from the authors of `SHARC`–v2.1 to distribute this modified version of the code, and we were given permission. We are grateful to the authors of `SHARC`–v2.1 for making their code available and for their cooperation every step of the way. Some of the modifications in `SHARC-MN` are scheduled to be included in a future version of `SHARC` itself.

Our work on the MN extension of `SHARC`–v2.1 was supported in part by the U. S. Department of Energy, Office of Basic Energy Sciences.

Citations

Users of `SHARC-MN`–1.1 are requested to cite both `SHARC` and `SHARC-MN`. Examples of appropriate referencing are as follows:


See also the references for methods in Sections I.D and I.E.
I. Introduction

SHARC-MN is an extended version of SHARC. Both codes are used for direct dynamics calculations of electronically nonadiabatic processes in which all needed energies, gradients, and nonadiabatic couplings (NACs) are calculated by performing electronic structure calculations as they are needed in the dynamics calculations.

When molecules and materials are electronically excited, the nuclear motion is affected. Processes involving more than one electronic state are called electronically nonadiabatic dynamics. The SHARC and SHARC-MN codes are used for semiclassical simulation of nonadiabatic dynamical processes, i.e., for computing where nuclei move and how electronic coefficients change and decohere. Within the semiclassical approach, one typically treats the nuclei as undergoing classical motion on an effective potential or as classical motion that switches between two or more potentials, while in either case the electrons are treated quantum mechanically with the nuclear and electronic propagations being coupled. There are two main categories of methods:

- trajectory surface hopping (TSH): Nuclei are propagated at any one time on a single potential energy surface (PES) that is created by electronic structure, and the PES is switched stochastically from time-to-time as the probability of being in different electronic states changes; this switch is called a “hop”. We refer the readers to the SHARC manual (https://sharc-md.org/?page_id=15) for detailed information about using SHARC or SHARC-MN for TSH calculations.
- methods based on a self-consistent potential (SCP): Nuclei are propagated on a mean-field PES. These methods are in SHARC-MN but not (at this time) in SHARC, and they are described in this manual. SHARC-MN-v1.1 includes three classes of methods of this type:
  - semiclassical Ehrenfest (SE),
  - coherent switching with decay of mixing (CSDM)
  - self-consistent decay of mixing (SCDM)

Note that that SE is not recommended for most practical applications because it does not produce physical final states. SCDM is an older version that CSDM and has been found to be less accurate in comparison to converged quantum dynamics. Therefore, although SHARC-MN contains all three methods, the one that is recommended is CSDM.

The nonadiabatic coupling vectors (NACs) that are usually used for direct dynamics do not conserve angular momentum or the position of the center of mass. SHARC-MN corrects this problem by using a projection operator to remove the translational and rotational components of the originally computed NACs or the effective NACs used in some of the methods.

The implementations of CSDM, SCDM, SE, and NAC projection and the incorporation of adaptive time steps are the first set of extensions of SHARC that are included in SHARC-MN.

The second set of extensions consists of adding the time-derivative versions of the SE, CSDM, and SCDM methods. These are called tSE, tCSDM, and tSCDM. Among these, tCSDM is recommended. The time-derivative modification allows one to use time derivatives rather than NACs; this is more convenient and more efficient, and it allows more accurate integrations in regions where the NACs show sharp spikes.

The third set of extensions consists of adding the curvature-driven versions of the SE, CSDM, and TSH methods. These are called kSE, kCSDM, and kTSH. Among these, kCSDM is recommended. The curvature-driven modification allows one to use curvatures of the adiabatic
potential energy surfaces along the nuclear-motion path instead of NACs or time derivatives. This is most convenient of all.

I.A. Introduction to semiclassical Ehrenfest and CSDM

Semiclassical Ehrenfest is similar to surface hopping in that the nuclear wave packet motion is approximated by classical trajectories. However, in contrast to surface hopping, the nuclei in semiclassical Ehrenfest dynamics are propagated on a self-consistent potential. One obtains the self-consistent potential by averaging the PESs over the electronic states considered with weights given by the electronic density matrix.

The advantages of semiclassical Ehrenfest dynamics are:

- The method is derived from the time-dependent Schrödinger equation by a self-consistent field approximation. Employing the self-consistent potential is a natural way to simulate the unitary (fully coherent) motion in which the wave function is a linear combination involving multiple electronic states.[1]
- Employing the self-consistent potential makes semiclassical Ehrenfest dynamics invariant with respect to the choice of electronic-state representation, which may be either adiabatic states or diabatic states.
- As in the original surface hopping method, the propagation of the classical trajectories only requires local information about the PESs and couplings, and the nuclear equations of motion follow Newtonian mechanics (although the motion is coupled to electronic motion in a way that has no classical analog).

However, semiclassical Ehrenfest dynamics is notoriously bad for long-time propagation as the trajectory may propagate on unphysical averaged PESs after leaving a strong interaction region, e.g., a region with locally avoided crossing of adiabatic PESs. This poor behavior is due to the lack of decoherence.[2,3,4] In molecules and materials, the electronic density matrix is a reduced density matrix for the electronic subsystem embedded in a “bath” of nuclei, and the nuclear motion causes decoherence of the electronic density matrix. Decoherence causes the density matrix to tend to a diagonal form in an environmentally selected basis called the pointer basis. The robustness of semiclassical Ehrenfest dynamics with respect to the choice of representation is important because in general one does not know which representation (adiabatic, diabatic, or something else) is closest to the pointer basis,[5,6] although one does know that the adiabatic basis is the pointer basis when the adiabatic approximation is a good approximation.

The CSDM method adds non-Markovian decoherence to the semiclassical Ehrenfest method so that the electronic coherences (i.e., the off-diagonal elements of the electronic density matrix) decay to zero in the assumed pointer basis after one leaves a region of strongly coupled potential energy surfaces.[4,7] SHARC-MN assumes that the pointer basis is the adiabatic one; this is a reasonable assumption for many cases. As a consequence, a CSDM trajectory propagates on a single PES in a pure adiabatic state in asymptotic regions or other regions far from strong interaction regions.

SHARC-MN can also do calculations by semiclassical decay of mixing (SCDM) [3]. The SCDM method, like SE and CSDM, is a self-consistent potential method, and it may be considered an older version of CSDM with a less accurate (but simpler) treatment of decoherence. We recommend CSDM over SCDM because it is more accurate, and therefore we de-emphasize SCDM in the manual. The user who is interested in SCDM (for historical reasons
or because it is simpler) should note, however, that any option that is available for CSDM is also available for SCDM.

I.B. Important note

To understand SHARC-MN, users must read the original SHARC manual. This manual only has descriptions of methods and keywords that are added to the SHARC code.
I.C.1 New features in *SHARC-MN*–v1.1 (additions to *SHARC*–v2.1):

These features are new in *SHARC-MN*–v1.0 (2020):

- **Dynamics program **`sharc.x`**:
  - New methods: semiclassical Ehrenfest (SE) dynamics, coherent switching with decay of mixing (CSDM), which adds non-Markovian decoherence to semiclassical Ehrenfest(SE), and self-consistent decay of mixing (SCDM, which is an older, less accurate version of CSDM).
  - Time-derivative versions of SE, CSDM, and SCDM dynamics based on overlap. These methods are called tSE, tCSDM, and tSCDM.
  - Adaptive time-step propagation: adaptive Velocity Verlet or Bulirsch-Stoer
  - Projected NAC for conserving nuclear angular momentum and the position of the center of mass in direct dynamics with TSH, SE, CSDM, SCDM, tSE, tCSDM, or tSCDM.

- **Data extraction**:
  - New options in `data_extactor.x`

- **Auxiliary scripts**:
  - `setup_traj.py` works for setting up both surface hopping and self-consistent potential methods, where the self-consistent potential method may be SE, CSDM, SCDM, tSE, tCSDM, or tSCDM.

I.C.2 New features in *SHARC-MN*–v1.1:

These features are new in *SHARC-MN*–v1.1 (2021):

- **Dynamics program **`sharc.x`**:
  - New methods: curvature-based approximation of time-derivative coupling. This is used to create a set of new methods, including curvature-driven trajectory surface hopping (κTSH), curvature-driven semiclassical Ehrenfest (κSE), and curvature-driven coherent switching with decay of mixing (κCSDM). These methods do not require electronic-structure calculations of nonadiabatic coupling vectors (NACs) or wave function overlaps. Therefore, κCSDM, κSE, and κTSH only require evaluation of adiabatic energies (potential energy surfaces) and their gradients at each time step.

- **Auxiliary scripts**:
  - `setup_traj.py` added the curvature-based approximation option for coupling, the keyword is “etdc”.

I.D. References: basic methods

The following references may be cited for the basic methods in *SHARC-MN*:


In addition, users may cite the references in section I.E for specific dynamical methods.
I.E. References: specific methods
- semiclassical Ehrenfest (SE) [8,9]
- coherent switching with decay of mixing (CSDM) [8]
- tSE and tCSDM: methods that use time-derivative couplings [10]
- Projected NACs [11]
- Curvature-driven methods, κCSDM, κSE, and κTSH [14]

I.F. Installation
Installation of *SHARC-MN* requires the system specific modification of **Makefile** that is located in the **source** folder. Installation is done by a single command from the **source** folder:

```
make
```

The *SHARC-MN* executable is **sharc.x**; the output data extractor is called **data_extractor.x**.
II. Input Keywords

II.A. List of keywords

Table II.1 gives keywords that are new or have new options in SHARC-MN as compared to SHARC. (The overlap keyword has not changed, but it is included for completeness.) Users should refer to Table 4.1 in original SHARC manual for other keywords.

Table II.1: Input keywords for sharc.x. The first column gives the name of the keyword, the second lists possible arguments, and the third line provides an explanation. Defaults are marked in green like this. $n$ denotes the n-th argument to the keyword.

<table>
<thead>
<tr>
<th>Keyword</th>
<th>Arguments</th>
<th>Explanation</th>
</tr>
</thead>
<tbody>
<tr>
<td>method</td>
<td>string</td>
<td>Set the simulation method.</td>
</tr>
<tr>
<td></td>
<td>$1=tsh$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$1=scp$, ehrenfest</td>
<td></td>
</tr>
<tr>
<td>nsubsteps</td>
<td>integer</td>
<td>Number of substeps for the integration of the electronic equation of motion. This is equal to the ratio of the step size for integrating the nuclear equations of motion to the step size for integrating the electronic equations. It is recommended to use a large number of substeps (e.g., 200) when performing SE and CSDM.</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td></td>
</tr>
<tr>
<td>coupling</td>
<td>string</td>
<td>Quantities describing the nonadiabatic couplings.</td>
</tr>
<tr>
<td></td>
<td>$1=ddr$, nacdr</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$2=ddt$, nacdt</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$3=overlap$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$4=etdc$</td>
<td></td>
</tr>
<tr>
<td>decoherence_scheme</td>
<td>string</td>
<td>Method for decoherence correction. No decoherence correction.</td>
</tr>
<tr>
<td></td>
<td>$1=none$</td>
<td>Energy-difference-based correction for TSH. The TSH/EDC scheme is explained in Refs. [12,13] and is based on an earlier approximation [4] to the decoherence time in terms of energy gaps and nuclear kinetic energies.</td>
</tr>
<tr>
<td></td>
<td>$1=afssh$</td>
<td>Augmented FSSH for TSH (refer to original SHARC manual for details of augmented FSSH)</td>
</tr>
<tr>
<td></td>
<td>$1=dom$</td>
<td>Add decay-of-mixing decoherence terms to SE to perform CSDM or SCDM.</td>
</tr>
<tr>
<td>switching_procedure</td>
<td>string</td>
<td>Method for scheme used to switch the pointer state in decay-of-mixing calculations</td>
</tr>
<tr>
<td></td>
<td>$1=csdm$</td>
<td>Self-consistent decay of mixing.[3,8]</td>
</tr>
<tr>
<td></td>
<td>$1=scdm$</td>
<td></td>
</tr>
</tbody>
</table>
**neom string**

$1=\text{ddr, nacdr}$

$1=\text{gdiff}$

Method for propagating nuclear equations of motion for self-consistent-potential methods (SE, CSDM, or SCDM)

Use full NAC (this is the default when coupling is set to ddr or nacdr)

Use effective NAC, which is a combination of difference gradient vector and velocity (this is the default when coupling is set to overlap)

**integrator string**

$1=\text{fvv}$

$1=\text{avv}$

$1=\text{bsh}$

Will dynamics be calculated with adaptive time step?

- Fixed time-step Velocity Verlet Integrator
- Adaptive time-step Velocity Verlet Integrator
- Bulirsch-Stoer Integrator

**convthre float**

$1=1e-04 \text{ eV}$

$1=1e-04 \text{ a.u.}$

Convergence threshold for successive steps

- Use adaptive Velocity-Verlet integrator.
- Use Bulirsch-Stoer integrator.

**dtmin float**

$0.0001 \text{ fs}$

Minimum time step allowed in adaptive Velocity Verlet in fs.

**nac_projection**

**nonac_projection**

Applies projected NAC (default)

Applies original NAC

---

**II.B. Detailed description of the keywords**

**method**

The **method** keyword controls the method to be used for dynamics

- **tsh** for trajectory surface hopping
- **scp** for methods with self-consistent potentials, which are SE, CSDM, and SCDM.

To run CSDM, which is recommended, set **method** to **scp** and **decoherence_scheme** to **dom**.

**coupling**

The **coupling** keyword controls the coupling used in propagation of electronic coefficients

- **edtc** curvature driven time derivative coupling,

\[
\sigma_{IJ} = \langle \psi_I | \partial_{\tilde{t}} | \psi_J \rangle \approx \frac{1}{2} \left[ \frac{\partial^2 (V_I - V_J)}{\partial \tilde{t}^2} \right]^{1/2}
\]

**integrator**

The **integrator** keyword controls the integrator to be used in dynamics

- **fvv** for fixed time step Velocity Verlet
- **avv** for adaptive time step Velocity Verlet,
- **bsh** for Bulirsch-Stoer integrator

The **fvv** integrator is same as the default integrator in previous *SHARC* programs. We recommend using **fvv** or **avv**.

**convthre**
The `convthre` keyword controls the maximum allowed differences between successive time steps in adaptive integrators. For `avv`, it is the maximum allowed energy difference in the current and previous time steps.

**dtmin**

The `dtmin` keyword controls the minimum allowed time step for adaptive integrators.

**decoherence_scheme**

When performing self-consistent potential based methods, the decoherence is introduced with the decay-of-mixing algorithm. Use of the decay-of-mixing decoherence scheme is turned on by setting `decoherence_scheme dom`. The other two options, i.e., `decoherence_scheme edc` and `decoherence_scheme afssh`, are used for calculations with the trajectory surface hopping method.

**switching procedure**

There are two options for the computation of the probabilities of switching pointer states in the decay-of-mixing algorithms, and they can be set with the `switching_procedure` keyword.

- `switching_procedure csdm` turns on coherent switching with decay of mixing [4,8], which is the default and is recommended.
- `switching_procedure scdm` turns on the older self-consistent decay of mixing [3,8]

**neom**

The `neom` keyword is used to control the form of the nonadiabatic coupling vector in the nuclear equations of motion; note that this is a separate choice from the form of the NAC used in the electronic equations of motion. One can directly use the nonadiabatic coupling vector by using either `neom nacdr` or `neom ddr`; this will require using an electronic structure theory code to compute the nonadiabatic coupling vector. Therefore, the nuclear equation of motion follows original format for SE and CSDM, i.e.,

\[
\left[ \dot{\mathbf{P}} \right]_{SE} = - \sum_{I} \text{Re}(\rho_{II}) \frac{\partial V_j}{\partial \mathbf{R}} + \sum_{I} \sum_{J \neq I} \text{Re}(\rho_{IJ})(V_I - V_J) \mathbf{d}_{IJ}
\]

\[
\left[ \dot{\mathbf{P}} \right]_{CSDM} = \left[ \dot{\mathbf{P}} \right]_{SE} + \left[ \dot{\mathbf{P}} \right]_{DM}
\]

\[
\left[ \dot{\mathbf{P}} \right]_{DM} = \sum_{I \neq K}^{N} \frac{\rho_{II}(t)(V_I - V_K)}{\tau_{IK}} \left( \mathbf{s}_{IK} \cdot \dot{\mathbf{v}} \right)
\]

Alternatively, one can use an effective nonadiabatic coupling vector by setting `neom gdiff`. The effective nonadiabatic coupling vector is a vector defined in Section IV.C as a combination of the difference gradient vector and the velocity vector. The dot product between the effective nonadiabatic coupling vector and velocity vector equals the overlap of electronic wave functions at successive steps. See reference [10] for detailed description of effective nonadiabatic coupling vector. Using the effective nonadiabatic coupling vector is the default option if one uses time-derivative coupling to propagate electronic coefficients, which is set by `coupling overlap`. If one uses `neom gdiff`, one must use `coupling overlap`, and if one uses `coupling overlap` one may...
use **neom nacdr**, **neom nacdr**, or **neom gdiff**. When using effective NAC in nuclear equation of motion, to conserve the energy of the system, the form has to be changed to,

\[
\mathbf{P}_{SE} = -\sum_I \text{Re}(\rho_I) \frac{\partial V_I}{\partial \mathbf{R}} + \sum_I \sum_{J \neq I} \left[ \frac{\text{Re}(\rho_{IJ})(V_I - V_J)\sigma_{IJ}}{\mathbf{v} \cdot \mathbf{G}_{IJ}} \right] \cdot \mathbf{G}_{IJ}
\]

where \( \mathbf{G}_{IJ} \) is effective NAC, and \( \sigma_{IJ} \) is time derivative coupling between states \( I \) and \( J \).

One does not need to set up **neom** keyword. In that situation, we will employ the default choice of nuclear EOM based on **coupling** keyword. The following are default maps between **coupling** and **neom**,

- \( \text{coupling=ddr, nacdr} : \text{neom=ddr, nacdr} \)
- \( \text{coupling=overlap} : \text{neom=gdiff} \)
- \( \text{coupling=etdc} : \text{neom=gdiff} \)

**nac_projection** and **nonac_projection**

These two keywords control the use of projected nonadiabatic coupling vector or original nonadiabatic coupling vector. The projected nonadiabatic coupling vector conserves nuclear angular momentum and the position of the center of mass, and it is the recommended choice; it is also the default. We refer readers to reference [11] for a detailed description of the projection.
II.C. Combinations of keywords

Possible combinations of the keywords are shown in Table II.2.

**Table II.2**: Possible combinations of `coupling`, `neom`, and `nac_projection` keywords for running SE or CSDM calculations\(^a\)

<table>
<thead>
<tr>
<th>keywords</th>
<th>nuclear EOM</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td><code>nardr(ddr)</code></td>
<td><code>nardr(ddr)</code></td>
<td>eq 21</td>
</tr>
<tr>
<td><code>nardr(ddr)</code></td>
<td><code>nardr(ddr)</code></td>
<td>eq 9</td>
</tr>
<tr>
<td><code>overlap</code></td>
<td><code>nardr(ddr)</code></td>
<td>eq 21</td>
</tr>
<tr>
<td><code>overlap</code></td>
<td><code>nacdr(ddr)</code></td>
<td>eq 9</td>
</tr>
<tr>
<td><code>overlap</code></td>
<td><code>gdiff</code></td>
<td>eq 10</td>
</tr>
<tr>
<td><code>overlap</code></td>
<td><code>gdiff</code></td>
<td>eq 17</td>
</tr>
<tr>
<td>`nonac_projection</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(^a\)All three keywords are used in all of the self-consistent-potential methods.

Note the following:
- “coupling” controls the equation of motion for electronic coefficients.
- “neom” controls the equation of motion for nuclei.

Originally, the electronic and nuclear equations of motion had a 1-to-1 map.
- `coupling=ddr` automatically assumed `neom=ddr`
- `coupling=overlap` automatically assumed `neom=gdiff` (which specifies using the effective NAC)
- `coupling=etdc` automatically assumed `neom=gdiff`

However, the current version of SHARC-MN allows possibilities to combine the options more flexibly. Most of these other possibilities would probably never be the method of choice with one possible exception, namely `coupling=overlap` and `neom=ddr`, which would use the TDC to propagate the electronic equation of motion and use the original NAC form of the nuclear equation of motion.

One does not have to set up `neom`; if it is not set up the program will use the automatic map mentioned above.

The sample input in Appendix B of the manual and the additional sample inputs in the Supporting Information of ref. [14] may be helpful in understanding the usage of the keywords.
III. Output

SHARC provides an auxiliary program data_extractor.x, which can operate on the output.dat file to extract properties as a function of time along the trajectory. We refer the users to Table 7.5 and Section 7.10 Data Extractor: data_extractor.x in original SHARC manual for a detailed description of the data_extractor.x program. Here we describe additional options for this program.

Table III.1. Additional command-line options for data_extractor.x.

<table>
<thead>
<tr>
<th>Option</th>
<th>Description</th>
<th>Default</th>
</tr>
</thead>
<tbody>
<tr>
<td>-h</td>
<td>Display help message and quit.</td>
<td>Help not displayed</td>
</tr>
<tr>
<td>-f</td>
<td>File name</td>
<td>None – file name must be given.</td>
</tr>
<tr>
<td>-sk</td>
<td>skip parsing of geom., vel., grad., NAC.</td>
<td>False</td>
</tr>
<tr>
<td>-ccd</td>
<td>Write ccoeff_diag.out</td>
<td>True</td>
</tr>
<tr>
<td>-dend</td>
<td>Write density_diag.out</td>
<td>False</td>
</tr>
<tr>
<td>-cdend</td>
<td>Write cdensity_diag.out</td>
<td>False</td>
</tr>
<tr>
<td>-denm</td>
<td>Write density_mch.out</td>
<td>False</td>
</tr>
<tr>
<td>-rdd</td>
<td>Write den_state_diag.out</td>
<td>False</td>
</tr>
<tr>
<td>-crdd</td>
<td>Write cdens_state_diag.out</td>
<td>False</td>
</tr>
<tr>
<td>-rdm</td>
<td>Write den_state_mch.out</td>
<td>False</td>
</tr>
<tr>
<td>-xs</td>
<td></td>
<td>Default</td>
</tr>
<tr>
<td></td>
<td>-e, -rdm, -rdd, -d, -cd, -cm, -p, -x</td>
<td></td>
</tr>
<tr>
<td>-s</td>
<td></td>
<td>-ccd, -sp, -xm, -cb, -da plus -xs</td>
</tr>
<tr>
<td>-l</td>
<td></td>
<td>-crdd, -id, -im, plus -s</td>
</tr>
<tr>
<td>-xl</td>
<td></td>
<td>-ccd, -dend, -cdend, -denm, plus -l</td>
</tr>
</tbody>
</table>
Table III.2. Content of the files written by `data_extractor.x` corresponding to the additional options shown in Table III.1.

- \( n \) is the total number of states.
- \( j \) is a state index \((j \in \{1, \ldots, n\})\).

<table>
<thead>
<tr>
<th>File</th>
<th>Columns</th>
<th>Columns</th>
</tr>
</thead>
<tbody>
<tr>
<td><code>ccoeff_diag.out</code></td>
<td>(2 + 2n)</td>
<td>1</td>
</tr>
</tbody>
</table>
|                     | 2       | Norm of coherent wave function:  
|                     | \[ \sum_j [c_{diag,j}^{\text{coherent}}]^2 \]  
|                     | \(1 + 2j\) | \(\text{Re}\left[ c_{diag,j}^{\text{coherent}} \right] \)  
|                     | \(2 + 2j\) | \(\text{Im}\left[ c_{diag,j}^{\text{coherent}} \right] \)  
| `density_diag.out`  | \(1+(n+1)*n/2\) | 1       | Time: \( t \) (fs) |
|                     | \(1+n*(j-1)+k-j+1\) | Density matrix element \(D_{ij}\) in the diagonal representation.  
| `cdensity_diag.out` | \(1+(n+1)*n/2\) | 1       | Time: \( t \) (fs) |
|                     | \(1+n*(j-1)+k-j+1\) | Coherent density matrix element \(\left[ D_{ij}^{\text{coherent}} \right]\) in the diagonal representation.  
| `density_mch.out`   | \(1+(n+1)*n/2\) | 1       | Time: \( t \) (fs) |
|                     | \(1+n*(j-1)+k-j+1\) | Density matrix element \(D_{ij}\) in MCH representation.  
| `den_state_diag.out`| \(1 + n\) | 1       | Time \( t \) (fs) |
|                     | \(n\) | Real diagonal density matrix element \(\text{Re}\left[ D_{ii}^{\text{coherent}} \right]\) in the diagonal representation.  
| `cden_state_diag.out`| \(1 + n\) | 1       | Time \( t \) (fs) |
|                     | \(n\) | Real diagonal coherent density matrix element \(\text{Re}\left[ D_{ii}^{\text{coherent}} \right]\) in the diagonal representation.  
| `den_state_mch.out` | \(1 + n\) | 1       | Time \( t \) (fs) |
|                     | \(n\) | Real diagonal density matrix element \(\text{Re}\left[ D_{ii}^{\text{coherent}} \right]\) in the MCH representation.  

IV. Theory

IV.A. Decoherence by decay of mixing

Decay of mixing is described in Refs. [2,3,4,8]. We refer the reader to Ref. [8] for details of implementing decay of mixing in a way compatible with SHARC; the decoherent propagator is introduced in the MCH representation. The decay-of-mixing decoherence scheme is used by decoherence_scheme dom. The electronic coefficients are propagated with the following electronic equation of motion:

$$c_{\text{diag}}(t + \Delta t) = U(t + \Delta t) \left[ P_C^{\text{MCH}}(t + \Delta t) P_D^{\text{MCH}}(t + \Delta t) \right] U(t) c_{\text{diag}}(t)$$

(1)

The coherent propagator and decoherent propagator (which is diagonal) are

$$P_C^{\text{MCH}}(t + \Delta t, t) = \exp \left( - \left( i H^{\text{MCH}} + \mathbf{v} \cdot \mathbf{d}^{\text{MCH}} \right) \Delta t \right)$$

(2)

$$\left[ P_D^{\text{MCH}}(t + \Delta t, t) \right]_{II} = \begin{cases} - \frac{\Delta t}{2 \tau_I(t)} & I \neq K \\ \sum_{j=K}^N \frac{\Delta t}{2 \tau_J(t)} \frac{\rho_{II}^{\text{MCH}}(t)}{\rho_{KK}^{\text{MCH}}(t)} & I = K \end{cases}$$

(3)

where $\tau_I$ is the decoherence time for state $I$:

$$\tau_I = \frac{\hbar}{|V_I - V_K|} \left( C + \sum_{i}^{N_{\text{atoms}}} \frac{2 E_0}{m_i} \right)$$

(4)

where $V_I$ is the adiabatic energy of state $I$, and $V_K$ is the adiabatic energy of state $K$. The default values of the parameters are $C = 1$ (unitless) and $E_0 = 0.1$ hartree. The decoherent direction $s_{IK}$ is given by,

$$s_{IK} = \text{Re} \left( \frac{a_0 P_{\text{vib}} \cdot d_{IK}}{|d_{IK}|} \right) + P_{\text{vib}}$$

(5)

where $a_0 \equiv 1$ bohr, $P_{\text{vib}}$ is the internal vibrational momentum (computed by removing the overall angular motion from the total momentum of the molecule), and $d_{IK}$ is the nonadiabatic coupling vector between state $I$ and decoherent state $K$. We use “decoherent state” and “pointer state” as synonyms.

IV.B. Pointer-state switching

The CSDM pointer-state switching probability between decoherent state $K$ and another state $I$ is:
where $\hat{c}_K^{\text{diag}}(t)$ are the coherent coefficients propagated fully coherently along the trajectory, and they are reset to $\hat{c}_K^{\text{diag}}(t)$ at each local minimum $D_K$. The coupling strength $D_K$ is defined differently, depending on which coupling is used. When using the nonadiabatic coupling vector,

$$D_K(t) = \sum_{l \neq K}^{N} |d_{IK}|$$  \hspace{1cm} (8)

When using the overlap,

$$D_K(t) = \sum_{l \neq K}^{N} |\sigma_{IK}|$$  \hspace{1cm} (9)

Using equation 9 is similar to the CSDM-C version in original CSDM reference[4]. The keyword coupling controls the use of equation 8 or 9 automatically; equation 8 corresponds to coupling ddr or coupling nacdr, and equation 9 corresponds to coupling overlap. The negative switching probabilities are set to zero and

$$P_{K \to K} = 1 - \sum_{l \neq K}^{N} P_{K \to l}$$  \hspace{1cm} (10)

We refer the reader to reference [8] for details. This is used with switching_PROCEDURE csdm.

IV.C. Effective nonadiabatic coupling
The nuclear equation of motion in self-consistent potential methods in its original form requires usage of the full nonadiabatic coupling vector $d_{IJ}$:

$$\dot{\mathbf{P}}(t) = -\sum_{I} \text{Re}(\rho_{II}) \frac{\partial V_I}{\partial \mathbf{R}} + \sum_{I \neq J} \sum_{J \neq I} \text{Re}(\rho_{IJ}) (V_I - V_J) \mathbf{d}_{IJ}$$  \hspace{1cm} (9)

One can, however, employ an effective nonadiabatic couplings, $\mathbf{G}_{IJ}$, to replace $\mathbf{d}_{IJ}$. Using of $\mathbf{G}_{IJ}$ requires eq 9 to be rewritten as,

$$\dot{\mathbf{P}} = -\sum_{I} \text{Re}(\rho_{II}) \frac{\partial V_I}{\partial \mathbf{R}} + \sum_{I \neq J} \sum_{J \neq I} \left[ \frac{\text{Re}(\rho_{IJ}) (V_I - V_J) \sigma_{IJ}}{\mathbf{v} \cdot \mathbf{G}_{IJ}} \right] \mathbf{G}_{IJ}$$  \hspace{1cm} (10)

Then $\mathbf{G}_{IJ}$ is defined as,

$$\mathbf{G}_{IJ} = \mathbf{g}_{IJ} + \alpha \mathbf{R}$$  \hspace{1cm} (11)

where $\alpha$ is a parameter defined as
A consequence of eq 12 is that
\[
\dot{\mathbf{R}}(t) \cdot \mathbf{G}_{IJ} (t) = \sigma_{IJ} (t)
\]  
(13)
where
\[
\sigma_{IJ} (t) = \dot{\mathbf{R}}(t) \cdot \mathbf{d}_{IJ} (t)
\]  
(14)
is the time-derivative coupling. This is related to the nonadiabatic coupling by
\[
\sigma_{IJ} (t) = \dot{\mathbf{R}}(t) \cdot \mathbf{d}_{IJ} (t)
\]  
(15)
Comparison of eq 13 to eq 15 justifies calling \( \mathbf{G}_{IJ} \) an effective nonadiabatic coupling. To conserve the angular momentum, one should use a projected effective nonadiabatic coupling,
\[
\mathbf{G}_{ij}^{Q} = (1 - \mathbf{Q}) \left( \mathbf{g}_{ij} + \alpha \dot{\mathbf{R}} \right)
\]  
(16)
where \( \mathbf{Q} \) is a projection operator that removes translational and rotational components. Using the projected effective nonadiabatic coupling results in the nuclear equation of motion becoming
\[
\dot{\mathbf{P}} = -\sum_{IJ} \text{Re} \left( \rho_{IJ} \right) \frac{\partial V_{ij}}{\partial \mathbf{r}} + \sum_{IJ} \sum_{J \neq I} \left[ \frac{\text{Re} (\rho_{IJ})(V_{ij} - V_{ji}) \sigma_{IJ}}{\mathbf{v} \cdot \mathbf{G}_{IJ}^{Q}} \right] \mathbf{G}_{ij}^{Q}
\]  
(17)
Setting \texttt{neom ddr} and \texttt{neom nacdr} uses eq 9 to propagate the nuclear equation of motion, while setting \texttt{neom gdiff} uses eq 10 or 17 to propagate the nuclear equation of motion, depending on whether one sets \texttt{nonac_projection} or \texttt{nac_projection}.

IV.D. Projection operator

To conserve both the nuclear angular momentum and the position of the center of mass in nonadiabatic dynamics trajectories with TSH or self-consistent potential methods, one should use the projected nonadiabatic coupling or the projected effective nonadiabatic coupling.

The projection operator is a \( 3N \times 3N \) matrix with elements
\[
Q_{ii',JI'} = \frac{1}{N} \delta_{ii'} + \sum_{\alpha, \beta, \alpha', \beta'} \sum_{\gamma, \gamma'} \sum_{\alpha, \beta} \sum_{\alpha', \beta'} \sum_{\gamma, \gamma'} \varepsilon_{\alpha \beta} R_{\alpha \beta} \left[ \mathbf{I}^{-1} \right]_{\beta \beta'} \sum_{\gamma, \gamma'} \sum_{\gamma', \gamma} \varepsilon_{\alpha' \beta' \gamma} R_{i' \gamma} \varepsilon_{i \gamma}
\]  
(18)
where indices \( i \) and \( i' \) label the nuclei and vary from 1 to \( N \), \( \alpha, \beta, \gamma, \alpha', \beta', \) and \( \gamma' \) take on the values \( x, y, \) and \( z \), \( \mathbf{I}^{-1} \) is the inverse of matrix \( \mathbf{I} \), matrix \( \mathbf{I} \) is same as moment of inertia matrix with all masses set to 1, and \( \varepsilon \) is the completely antisymmetric third-order unit pseudotensor, whose elements are the Levi-Civita symbol. The first term of the projection operator projects onto the three directions corresponding to overall translation, and the second term projects onto the three directions corresponding to overall rotation.

Applying the projection operator to the nonadiabatic coupling results in a projected nonadiabatic coupling:
\[
\mathbf{d}_{ij}^{Q} = (1 - \mathbf{Q}) \mathbf{d}_{ij}
\]  
(19)
For TSH, one uses the projected NAC when the momentum after a hop is adjusted,
\[
\mathbf{P}_{i \text{post-hop}} = \mathbf{P}_{i \text{pre-hop}} - \alpha_{ij} \mathbf{d}_{ij}^{Q}
\]  
(20)
For semiclassical Ehrenfest and CSDM dynamics, one employs a self-consistent potential (SCP), and this yields
\[ \dot{P} = -\sum_{I=1}^{n} \text{Re} \left( \rho_{II} \right) \frac{\partial V_I}{\partial R} + \sum_{I=1}^{n} \sum_{J \neq I}^{n} \left[ \text{Re} \left( \rho_{IJ} \right) \left( V_I - V_J \right) \frac{\left( \textbf{v} \cdot \textbf{d}_{IJ} \right)}{\text{v} \cdot \text{d}_{IJ}^Q} \right] \text{d}_{IJ}^Q \]  
(21)

For CSDM, the decoherent direction is,
\[ \textbf{s}_{AK} = \frac{d_{0} \textbf{P}_{\text{vib}} \cdot \textbf{d}_{AK}^Q}{|\text{d}_{AK}^Q|} \text{d}_{AK}^Q + \textbf{P}_{\text{vib}} \]  
(22)

We refer users to reference [11] for more details.

IV.D. Curvature-driven time-derivative coupling

Setting the coupling keyword to etdc allows one employs an approximation to time derivative coupling that is computed from electronic wave function overlap integrals of successive time steps. This new approximation of time derivative coupling is called curvature driven time derivative coupling, $\kappa$TDC. Methods that use $\kappa$TDC in equation of motion are the series of $\kappa$ methods that described above, namely, $\kappa$SE, $\kappa$CSDM, and $\kappa$TSH. The $\kappa$TDC writes,
\[ \kappa\text{TDC} \equiv \sigma_{JI}^{\kappa} = \left( \phi_I | \frac{d}{dt} | \phi_J \right) \frac{1}{2} \left[ \frac{d^2 \left( V_I - V_J \right)}{dt^2} \frac{1}{V_I - V_J} \right]^{1/2} \]  
(23)

where $(V_I - V_J)$ is the local gap between adiabatic potential surfaces, and we use $\kappa$ as a prefix and as a superscript to denote approximations based on the curvature of the gap. Since the NAC is skew-Hermitian, we have
\[ \sigma_{JI}^{\kappa} = -\sigma_{IJ}^{\kappa} \]  
(24)

Equations 5 and 6 define the $\kappa$TDC. In practical implementation, we use
\[ \sigma_{JI}^{\kappa} \left( t + \Delta t \right) \approx \frac{1}{2} \left[ \frac{\Delta \dot{V}_{JI} \left( t + \Delta t \right) - \Delta \dot{V}_{JI} \left( t \right)}{\Delta t} \frac{1}{V_I - V_J} \right]^{1/2} \]  
(25)

where
\[ \Delta \dot{V}_{JI} \left( t \right) = \frac{\partial V_J \left( t \right)}{\partial R} \cdot \dot{\textbf{R}}(t) - \frac{\partial V_I \left( t \right)}{\partial R} \cdot \dot{\textbf{R}}(t) \]  
(26)

Because $\dot{\textbf{R}}(t + \Delta t)$ is not known at the stage of evaluation $\sigma_{JI}^{\kappa} \left( t + \Delta t \right)$, it is approximated by forward propagation,
\[ \dot{\textbf{R}}(t + \Delta t) \approx \dot{\textbf{R}}(t) + \textbf{a}(t) \Delta t \]  
(27)

where $\textbf{a}(t)$ is acceleration vector at time $t$. 
V. References


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Appendix B. Example Input

B.I. Example input for κCSDM

printlevel   0
geomfile     "geom"
veloc        external
velocfile    "veloc"

nstates      3 0 0
actstates    3 0 0
state        2 diag
coeff        auto
rngseed      5895967

ezero        -78.054411630961
tmax         250.0
integrator   fvv
stepsize     0.1
nsubsteps    200

method       scp
surf         diagonal
coupling     etdc
neom         gdiff
nogradcorrect
decoherence_scheme dom
B.II. Example input for \( \kappa \)TSH

```
printlevel  0
geomfile    "geom"
veloc       external
velocfile   "veloc"

nstates     3 0 0
actstates   3 0 0
state       2 diag
coeff       auto
rngseed      5895967

ezero       -78.054411630961
tmax        250.0
integrator  fvv
stepsize    0.1
nsubsteps   200

method      tsh
surf        diagonal
coupling    etdc
ekincorrect parallel_diff
decoherence_scheme edc
nogradcorrect

grad_sele
```