ACE Documentation

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1 Introduction

This document describes how to use the C++ code ACE for the solution of open quantum systems using the *automated compression of environments* (ACE) method. The article explaining the method can be found here.

ACE enables numerically exact simulations of the dynamics of an open quantum systems described by the quantum Liouville equation

$$\frac{\partial}{\partial t}\rho = -\frac{i}{\hbar}[H,\rho] + \mathcal{L}_{\text{nonh.}}[\rho] \tag{1}$$

where the microscopic Hamiltonian

$$H = H_S + H_E = H_S + \sum_{k=1}^{N_E} H_E^k, \tag{2}$$

is split up into system H_S and environment Hamiltonians H_E. The environment Hamiltonian H_E, which we define as also including the system-environment coupling, is assumed to be separable into N_E independent modes k. $\mathcal{L}_{nonh.}[\rho] = \mathcal{L}_S[\rho] + \mathcal{L}_E[\rho] = \mathcal{L}_S[\rho] + \sum_k \mathcal{L}_E^k[\rho]$ denotes non-Hamiltonian contributions to the dynamics such as Lindblad terms affecting the system and the environment.

The goal is to obtain the reduced system density matrix discretized on a time grid $t_l = t_a + l\Delta t$ up to a given final time $t_n = t_a + n\Delta t = t_e$. This can be done using the path integral expression

$$\rho_{\alpha_n} = \sum_{\substack{\alpha_{n-1}\dots\alpha_0\\ \tilde\alpha_n\dots\tilde\alpha_1}} \mathcal{I}^{(\alpha_n\tilde\alpha_n)\dots(\alpha_1\tilde\alpha_1)}\bigg(\prod_{l=1}^{n}\mathcal{M}^{\tilde\alpha_l\alpha_{l-1}}\bigg)\rho_{\alpha_0}, \tag{3}$$

where $\rho_{\alpha_l} = \rho_{\nu_l\mu_l}$ is the reduced system density matrix at time step l, $\mathcal{M} = \exp(-(i/\hbar)[H_S,\,.\,]\Delta t + \mathcal{L}_S[\,.\,]\Delta t)$ describes the free time evolution of the system without the environment, and $\mathcal{I}$ is the *process tensor* (PT) accounting for the effects of the environment. To keep the notation compact, we combine two Hilbert space indices on the system density matrix ν_l and μ_l into a single Liouville space index $\alpha_l = (\nu_l, \mu_l)$.

The PT can always expressed in the form of a matrix product operator (MPO)

$$\mathcal{I}^{(\alpha_n,\tilde\alpha_n)(\alpha_{n-1},\tilde\alpha_{n-1})\dots(\alpha_1,\tilde\alpha_1)} = \sum_{d_{n-1}\dots d_1} \mathcal{Q}^{(\alpha_n,\tilde\alpha_n)}_{1d_{n-1}}\mathcal{Q}^{(\alpha_{n-1},\tilde\alpha_{n-1})}_{d_{n-1}d_{n-2}}\dots\mathcal{Q}^{(\alpha_1,\tilde\alpha_1)}_{d_1 1}. \tag{4}$$

In the explicit derivation of the matrices $\mathcal{Q}$, the inner indices d_l correspond to a complete basis of the Liouville space of the full environment, whose dimension χ_l is typically extremely large. However, for MPOs, compression techniques are available, which systematically reduce their inner bonds, keeping only “relevant” parts of the environment influence while discarding "irrelevant" parts. A standard compression technique is to employ singular value decomposition (SVD), where the magnitude of the singular values determines the "relevance" of an environment degree of freedom. I.e., environment degrees of freedom corresponding to singular values σ_i below a predefined threshold $\sigma_i < \epsilon\sigma_0$ are disregarded, where σ_0 is the largest singular value.

The overall procedure can be understood as compressing the full environment propagator to its relevant subspace via a lossy transformation

$$\mathcal{Q}^{(\alpha_l,\tilde{\alpha}_l)}_{d_l,d_{l-1}} = \sum_{d'_l,d''_{l-1}} \mathcal{T}_{d_l,d'_l}\Big(e^{-(i/\hbar)[H_E,\,.\,]\Delta t+\mathcal{L}_E[\,.\,]\Delta t}\Big)_{d'_l,d''_l} \mathcal{T}^{-1}_{d''_{l-1},d_{l-1}}, \tag{5}$$

where the lossy transformation $\mathcal{T}$ and its pseudo-inverse $\mathcal{T}^{-1}$ are identified implicitly and automatically by the compression method. Hence the name: *Automated Compression of Environments*.

The main technical achievement is that the non-local environment influence is mapped onto a local one on an extended space, i.e., the system Liouville subspace extended by the inner dimension of the PT-MPO. Concretely, the reduced system density matrix is obtained by a sequence of matrix multiplications

$$\rho_{\alpha_l} = q_{d_l}\big(\mathcal{Q}^{(\alpha_l,\tilde{\alpha}_l)}_{d_l,d_{l-1}}\mathcal{M}^{\tilde{\alpha}_l,\alpha_{l-1}}\big)\dots\big(\mathcal{Q}^{(\alpha_2,\tilde{\alpha}_2)}_{d_2,d_1}\mathcal{M}^{\tilde{\alpha}_2,\alpha_1}\big)\big(\mathcal{Q}^{(\alpha_1,\tilde{\alpha}_1)}_{d_1,1}\mathcal{M}^{\tilde{\alpha}_1,\alpha_0}\big)\rho_{\alpha_0}, \tag{6}$$

where a summation over pairs of d_l, α_l, and $\tilde{\alpha}_l$ are implied, and the closure q_{d_l} traces out the environment, so that the reduced system density matrix can also be obtained at intermediate time steps $t_l < t_n$.

This description provides the conceptional basis for the ACE code. Some further points are worth highlighting:

- The code can be divided into three parts: Calculation of the PT-MPO, calculation of the system propagator $\mathcal{M}$, and the propagation via Eq.(equation6).

- PT-MPOs can be precalculated, stored in files, and reused. This facilitates the efficient sampling of different system Hamiltonians, e.g., corresponding to different system parameters of driving protocols.

- It is straightforward to use time-dependent system Hamiltonians, which can be read from input files.

- There are a number of convergence parameters that have to be set, such as the initial `ta` and final `te` time of the simulation, the time step `dt`, as well as MPO compression parameters like the threshold `epsilon` and discretization parameter `..._N_modes`. These have to be chosen carefully to ensure both, physical results and finite computation times.

- There are several ways to calculate PT-MPOs: The ACE algorithm is available for general environments composed of non-interacting environment modes. One can specify either the environment Hamiltonians for each mode seperately or use predefined generators for certain classes of environments. The algorithm by Jørgensen and Pollock as well as divide-and-conquer and periodic PT-MPOs are available for generalized spin-boson environments. Where they are available, they tend to perform much better than the brute-force solution using ACE.

- Multiple PT-MPOs can be used simultaneously, which provides a way to numerically exactly treat multi-environment problem. This is achieved by multiplying, in a single propagation step, with matrices $\big(\mathcal{P}^{(\alpha_l,\alpha'_l)}_{e_l,e_{l-1}}\mathcal{Q}^{(\alpha'_l,\tilde{\alpha}_l)}_{d_l,d_{l-1}}\mathcal{M}^{\tilde{\alpha}_l,\alpha_{l-1}}\big)$. Note, however, that for the matrix multiplication the inner dimensions of the different PTs are multiplied, which leads to exponential scaling with respect to the number of environments (and subsystems). There is work in progress to reduce these demands but, for now, this makes it impractical to propagate systems with more than 2 environments.

- The Trotter (time discretization) error can be reduced by employing symmetric Trotter decompositions. For a multi-environment problem, the order of matrix multiplications is alternated, i.e., $\big(\mathcal{P}^{(\alpha_l,\alpha'_l)}_{e_l,e_{l-1}}\mathcal{Q}^{(\alpha'_l,\tilde{\alpha}_l)}_{d_l,d_{l-1}}\mathcal{M}^{\tilde{\alpha}_l,\alpha_{l-1}}\big)$ for even time steps and $\big(\mathcal{M}^{\tilde{\alpha}_l,\alpha'_l}\mathcal{Q}^{(\alpha'_l,\tilde{\alpha}_l)}_{d_l,d_{l-1}}\mathcal{P}^{(\tilde{\alpha}_l,\alpha_{l-1})}_{e_l,e_{l-1}}\big)$ for odd time steps (we start counting from 0). Then, the time discretization error at even time steps is $\mathcal{O}(\Delta t^2)$ while there is an error $\mathcal{O}(\Delta t)$ at odd time steps. If there is only one environment, one can use a symmetric system-environment splitting $\big(\sqrt{\mathcal{M}}^{\tilde{\alpha}_l,\alpha'_l}\mathcal{Q}^{(\alpha'_l,\tilde{\alpha}_l)}_{d_l,d_{l-1}}\sqrt{\mathcal{M}}^{(\tilde{\alpha}_l,\alpha_{l-1})}\big)$, where $\sqrt{\mathcal{M}}$ is the system propagator over half a time step (to this end, set parameter `use_symmetric_Trotter` to `true`).

- There is work in progress to extract environment observables via inner bonds of PT-MPOs. To this end, the closures q_l are replaced by observable closures o_l, which have to be known ahead of the PT-MPO calculation.

2 Code, Compilation, Dependencies, and Design Choices

The code is written in C++ to combine low-level optimization (memory storage, access to LAPACK routines) with high-level abstraction. It has been most thoroughly tested on the Linux operating system but it also compiles and runs on Mac and Windows.

We have tried to keep the dependencies on other codes minimal. The Eigen library is very handy and provides useful and efficient routines, e.g., for matrix exponentials, so we make use of it at multiple places in the code. This dependency is harmless, as Eigen is "header only", i.e. it does not require installation beyond downloading the header files. However, there have been issues with matrix exponentiations in Eigen versions below 3.4, so it is strongly recommended to download the lates stable release.

The numerically most demanding part of ACE is the calculation of SVDs, for which LAPACK routines are available (optional), e.g., within the Intel MKL. These routines provide some degree of parallelization and, according to a few tests, reduce the computation time typically by a factor of 2-5. Note, however, using LAPACK routines, we have encountered instabilities for very small threshold values ($\epsilon \ll 10^{-12}$). Whether or not to use these routines is decided by the user at the compilation stage.

On Linux or Mac, Makefiles can be used for the compilation and some tuning can be done by command line arguments. For example, for the compiler to find the Eigen library, please set the variable `EIGEN_HOME` manually (only required for the compilation step) in such a way that the file `$EIGEN_HOME/Eigen/Eigen` exists. If the variable is not set, the script will try the most common directory `/usr/include/eigen3/`, which is, e.g., the place where Eigen is installed on Ubuntu Linux by the command `sudo apt install libeigen3-dev`.

If the MKL is installed on your system, the `MKLROOT` environment variable should be set to the correct directory. If this variable is set at the compilation stage, ACE will use the corresponding SVD routines.

To compile the code, set the corresponding environment variables, go into the main directory of ACE and type in the console

```
> make
```

This compiles the code and moves the binaries into the `bin/` subdirectory. For easy access later on, we suggest to add this directory to your Linux environment via the `PATH` variable. For example, add the follwing line to your ~`/.bashrc` file

```
PATH=/.../ACE/bin/:$PATH
```

where ... is to be replaced by the correct absolute path to the ACE directory. More programs, e.g., to post-process output data or manipulate PT-MPOs stored in files, can be compiled by running "`make tools`" from the ACE directory, which creates binaries in `/.../ACE/tools/`, which should then also be added to the path: `PATH=/.../ACE/bin/:/.../ACE/tools/:$P`

Log out and log in again to activate the changes. Then, go to a temporary directory and run

```
> ACE
```

This should generate a file `ACE.out` whose first lines are

```
0 0 0 1 0 0 0
0.01 0 0 1 0 0 0
0.02 0 0 1 0 0 0
0.03 0 0 1 0 0 0
0.04 0 0 1 0 0 0
...
```

Congratulations! You have just executed your first (rather boring) simulation using ACE.

Compilation on Windows has been tested using the GCC compiler in MinGW. We provide a compile script with `compile.bat` in the ACE directory. Please take a look and modify the path to the Eigen library according to your system specifics. Alternatively, install Windows Subsystem for Linux (WSL).

3 General usage

ACE is designed to be fully controllable by command line parameters, so the source code does not have to be modified and no prior knowledge in C++ required. Alternatively, the command line parameters can be composed into a driver file, which can then be passed to the code via the `-driver` command line option or simply as the first command line argument. Practitioners of scripting languages like Python may want to generate driver files using script, then run the programm, and then post-process the resulting output file.

Initial states, Hamiltonians, Lindblad terms, and observables for the system as well as for individual environment modes can be specified directly and quite generally, but additional sets of parameters are implemented that allow a more convenient control of certain common classes of environments.

Generally, times, energies, and temperatures are expected to be provided in units of ps, meV, and K, but it's easy to work with dimensionless quantities by rescaling the total Hamiltonian as well as the time, which are related by $\frac{\partial}{\partial(t/\lambda)}|\psi\rangle = (\lambda H)|\psi\rangle$.

Many objects, such as initial states, Hamiltonians, or observables are complex matrices. The ACE code has a rudimentary parser that interprets "matrix valued expressions" which are identified by curly braces. The bra-ket notation (or rather ket-bra expressions) is supported. Note that to generate the corresponding matrices, the Hilbert space dimension has to be specified. Concretely, `{|i><j|_d}` is interpreted as the operator that describes a transition from state j to state i in a d-dimensional space, where we use the convention that $i, j = 0, 1, \dots, d-1$.

For example, the Pauli matrix σ_y can be expressed as `{i*( -1*|1><0|_2 + |0><1|_2 )}`. Some operators are also known by name, such as the Pauli matrices `sigma_x`, `sigma_y`, `sigma_z`, or the identity matrix `Id_d`, where `d` has the be replaced by the respective dimension of the Hilbert space. Also supported are bosonic creation, annihilation, and number operators `bdagger_d`, `b_d`, and `n_d`, and direct products of matrices can be defined using `otimes` as in `{ hbar*1*(|0><1|_2 otimes bdagger_4 + |1><0|_2 otimes b_4)}`, which describes the Jaynes-Cummings interaction with coupling strength 1 between a TLS and a bosonic mode containing up to (4-1)=3 photons. Furthermore, some constants like `hbar` (in units of meV ps) or `pi` can be used as well as the square root function `sqrt(...)`.

Note that when specified directly on the command line, quotes are required around the curly braces to avoid interpretation of the curly braces by the `bash` shell. No quotes are used in parameter files.

To test if an operator-valued expression is well formed, you can use the tool `readexpression`, e.g., compare the outputs of

```
readexpression "{i*( -1*|1><0|_2 + |0><1|_2 )}"
```

with that of

```
readexpression "{sigma_y}"
```

Initial system states are given in terms of the initial system density matrix, which is specified by the parameter `initial`, which expects a matrix-valued expression in curly braces as described above. To extract relevant information, the values of a set of observables for each time step are written into an output file (file name specified by parameter `outfile`, default value: "ACE.out"). These observables are described by operators on the system Hilbert space, e.g., $\langle \hat{A} \rangle = \mathrm{Tr}_S(\hat{A}\rho_S)$, where ρ_S is the reduced system density matrix. The operators $\hat{A}$ have to be provided as matrix valued expressions to the parameter `add_Output`. This parameter can occur multiple times, with every occurrence adding another two columns to the output file, corresponding to real and imaginary parts of the respective operator average. If no parameter `add_Output` is specified, default values are used which are equivalent to the command line `-add_Output "{|1><1|_2}" -add_Output "{|0><0|_2}" -add_Output "{|0><1|_2}"`. This implies that it is absolutely necessary to specify `add_Output` explicitly when dealing with a system with more than two levels.

It is generally advised to specify the three parts—initial density matrix, system Hamiltonian, and output observables—in every parameter file. Of course, the dimensions have to be the same of all three types of matrices.

A few hands-on examples demonstrating the usage in concrete applications are given in the next sections.

4 Free system dynamics

In this section, we focus on the usage of the ACE code on examples without explicit environment parts, i.e., closed few-level systems or Lindblad master equations.

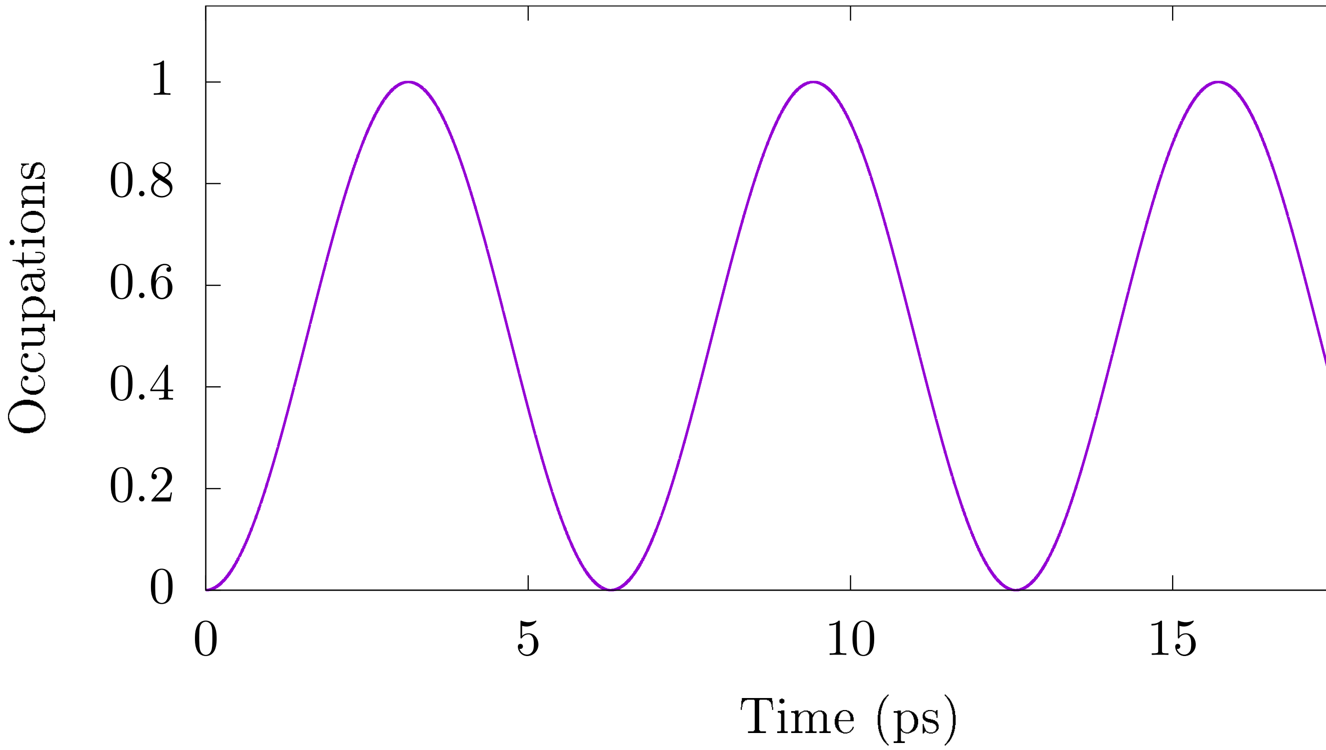
We begin with some of the most important parameters: The starting time, the final time, and the time step width can be specified by the command line options `-ta`, `-te`, and `-dt`, respectively, which have the default values 0, 10, and 0.01. You will find the corresponding time grid in the first column of output file `ACE.out`, whose name may be changed via the option `-outfile`. By default, there will be no environment, the system is a two-level system (TLS) initially in its ground state, and the system Hamiltonian is $H_S = 0$. For TLSs, if not specified otherwise, the second and third columns in the output file will be the real and imaginary part of the diagonal element of the system density matrix corresponding to the exited state. If no parameters are specified explicitly, these columns should remain 0.

As a first example, run

```
> ACE -dt 0.001 -te 20 -add_Hamiltonian "{hbar/2*(|1><0|_2+|0><1|_2)}" -outfile ACE1.out
```

This will generate an output file `ACE1.out`, which contains the dynamics of a constantly driven TLS from 0 to 20 ps with time steps of 0.001 ps. The driving is described by the Hamiltonian $H_S = (\hbar/2)(|X\rangle\langle G| + |G\rangle\langle X|)$ (note: hbar is given in units of meVps).

Plotting the second column of `ACE1.out` (in gnuplot: `plot "ACE1.out" using 1:2 with lines`) reveals clear Rabi oscillations of the excited state occupations:



The same result can be obtained creating and editing the file `driver1.param`:

```
dt                     0.001
te                    20

add_Hamiltonian        {hbar/2*(|1><0|_2+|0><1|_2)}


outfile                ACE1.out
```

and running

```
> ACE -driver driver1.param
```

or simply

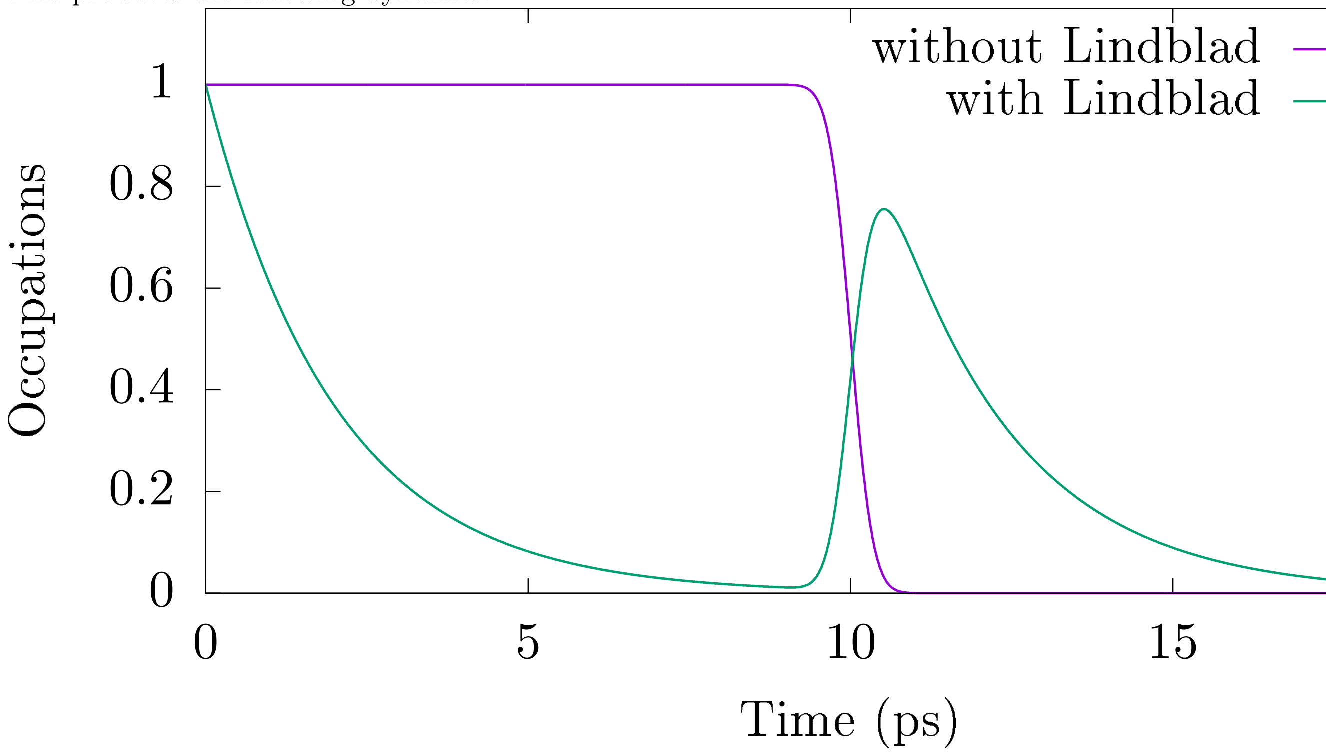
```
> ACE driver1.param
```

I.e., the first parameter is interpreted as a driver file.

A more complicated scenario can be described by the following driver file (`driver2.param`), where an initially excited TLS, optionally subject to radiative decay described by a Lindblad term, is driven by a Gaussian laser pulse:

```
dt                      0.01
te                     20

initial                {|1>&lt;1|_2}

#add_Lindblad            0.5  {|0>&lt;1|_2}
add_Pulse               Gauss  10 1 {1.*pi} 0  {hbar/2*|1>&lt;0|_2}

outfile                 ACE2.out
```

This produces the following dynamics:



where the two curves are results of calculations where the Lindblad term is either turned off or on. The # symbol in a driver file indicates a comment, i.e. anything after it is ignored. The parameters of `add_Lindblad` are the rate γ and the operator A for the Lindblad term

$$\gamma\mathcal{L}[A](\rho) = \gamma\Big[A\rho A^\dagger - \tfrac{1}{2}\big(A^\dagger A\rho + \rho A^\dagger A\big)\Big]. \tag{7}$$

The parameters of `add_Pulse Gauss` are the pulse center (here: $t_c = 10$ ps), the pulse duration ($\tau_{FWHM} = 1$ ps), the pulse area ($A = 1\pi$), the detuning ($\delta = 0$ meV), and the operator ($\hat{d} = \frac{\hbar}{2}|1\rangle\langle 0|$) describing the light-matter coupling, which enter the driving

Hamiltonian

$$H_D = \left(f(t)\hat{d} + f^*(t)\hat{d}^\dagger\right) \text{ with } f(t) = \frac{A}{\sqrt{2\pi}\sigma} e^{-\frac{1}{2}\frac{(t-t_c)^2}{\sigma^2}} e^{-i(\delta/\hbar)t}, \quad \sigma = \tau_{FWHM}/\sqrt{8\ln(2)}. \tag{8}$$

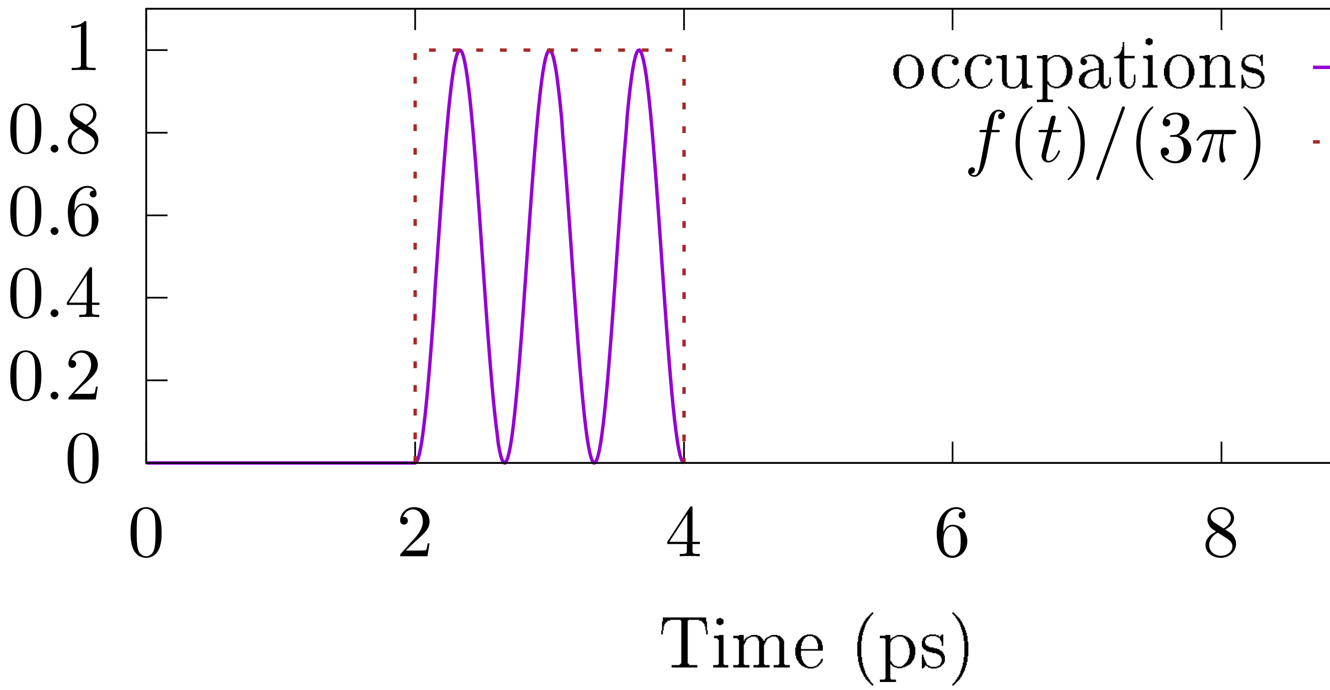
Alternatively, one can use arbitrary pulses by reading them from file. The first column is the time, the second and third columns are the real and imaginary parts of $f(t)$. For example, the file `step.pulse`, which contains

```
1.999999  0 0
2         1 0
3.999999  1 0
4.        0 0
```

As data points are linearly interpolated and the value of the first and last data points are extended to $-\infty$ and ∞, respectively, this describes a function whose real part is a rectangular function with height 1 from time 2 to 4 while the imaginary part is zero. Running `ACE` with the following driver file `pulse_from_file.param`

```
te          10
dt           0.01

initial    {|0><0|_2}

add_Pulse File step.pulse {(3*pi)*hbar/2*|1><0|_2}

add_Output {|1><1|_2}

outfile    pulse_from_file.out
```

Using the type `File` as the first argument of the parameter `add_Pulse` tells the code to read the file whose name is given as the second argument. The third argument is again the operator with which it enters the Hamiltonian. Note that the factor `3*pi` in the operator scales the pulse from height 1 to 3π, and because the pulse is finite for a time 2, the overall pulse area is 6π, i.e. 3 complete Rabi flops. This is clearly seen in the dynamics in the figure below



5 Including environments

In the following subsections, we demonstrate how environment can be included. First of all, note that as we include multiple environment modes, the total environment Liouville space becomes prohibitively large very fast and needs to be compressed to remain tractable, which is the core of the ACE algorithm. To enable compression, one has to specify a criterion. Most commonly, we use sweeps of singular value decompositions (SVDs) and only keep the subspaces related to singular values larger than a threshold $\epsilon\sigma_0$, where σ_0 is the largest singular value. This is enabled in the code by specifying a value to the paramter `threshold`. To provide small values, the C/C++ notation for powers of ten is useful, e.g., use `-threshold 1e-7` in the command line to enable compression with threshold $\epsilon = 10^{-7}$. The smaller the threshold, the more accurate the simulation. However, for very small thresholds also the calculation times as well as the memory demands increase. Alternatively, one can fix the maximal inner dimension of the PT MPO by defining `compress_maxk`. If both are specified, `compress_maxk` acts as an upper bound for the inner dimension.

In principle, all that is required to add a single mode is the environment Hamiltonian H_E including the interaction with the system and the initial environment mode density matrix, which can be provided in the form of two matrix valued expressions to the `add_single_mode` parameter. For example,

---

```
te          10
dt           0.01
```

```
threshold    1e-7

add_single_mode    {hbar*5* (Id_2 otimes n_3) + hbar*1.*(|0><1|_2 otimes bdagger_3 + |1><0


initial          {|1><1|_2}
add_Hamiltonian  { 0*Id_2 }
add_Output       {|1><1|_2}

outfile      singlemode.out
```

describes the Jaynes-Cummings coupling of a two-level system with a bosonic mode which is detuned with respect to the two-level system energy by a frequency 5 in units of the system-boson coupling (which is set to 1). This interaction conserves the number of excitations and the dynamics will look like strongly detuned coherent Rabi oscillations. Note that because H_E contains the system-environment interaction, it lives on the product Hilbert space of system and environment (system part comes first if `otimes` is used), whereas the environment initial density matrix lives in the bare environment Hilbert space.

Adding more modes is as simple as adding more `add_single_mode` lines to the parameter file. However, for many practically relevant bath more convenient sets of command line parameters are provided, some of which we discuss in the following subsection.

5.1 Fermionic environment

One of the predefined environments is defined by the Fermionic hopping Hamiltonian

$$H_E^k = \hbar g\big(c_k^\dagger c_S + c_S^\dagger c_k\big) + \hbar\omega_k c_k^\dagger c_k \tag{9}$$

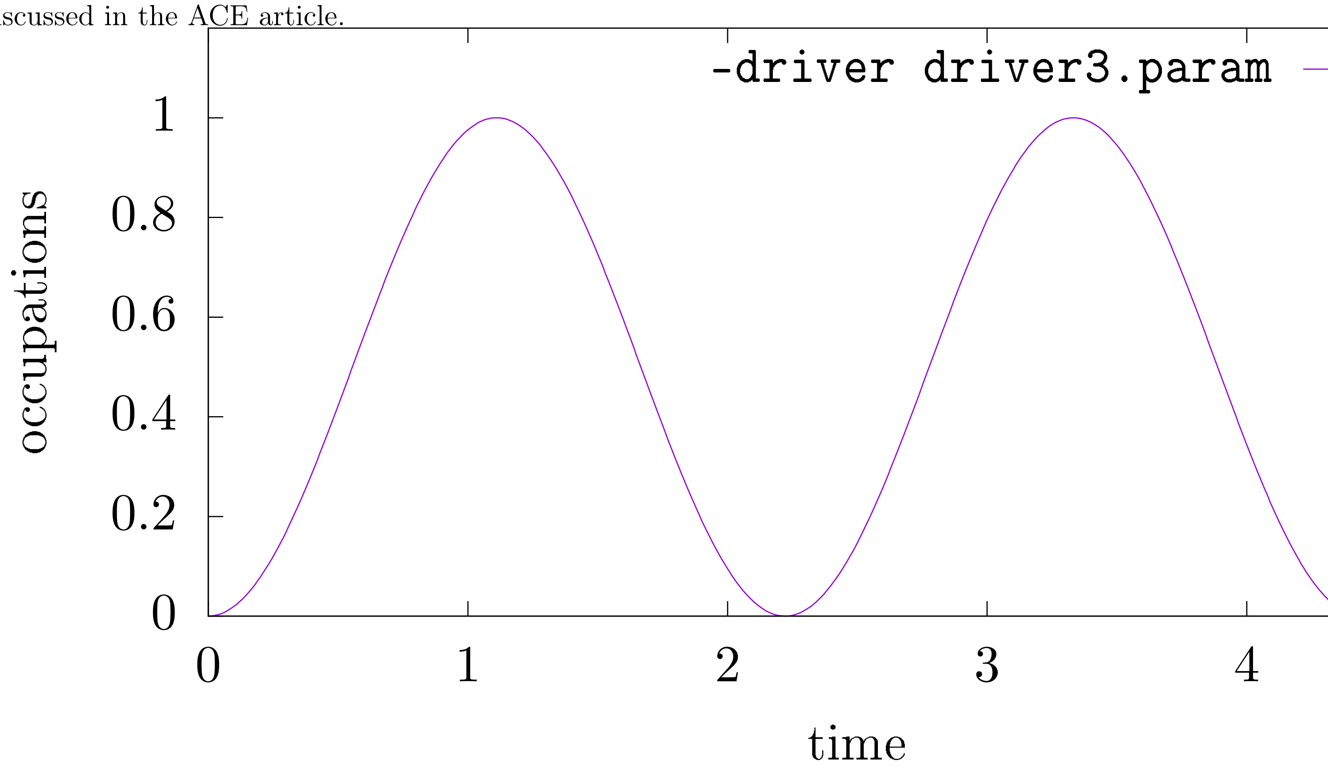
Here, the system is a Fermionic state that may be occupied $|1\rangle = c_S^\dagger|0\rangle$ or not $|0\rangle$. The occupation is created by $c_S^\dagger$ or destroyed by c_S. The environment consists of several Fermionic states, whose occupations are created and destroyed by $c_k^\dagger$ and c_k, respectively. In the limit $N_E \to \infty$, the environment consists of a continuum of state, which can be used to model the electronic states in metallic contacts in proximity to a molecule or a quantum dot. Consider the driver file:

```
te                  5
dt                  1e-2
threshold           1e-7

Fermion_N_modes       2
Fermion_g             1
Fermion_omega_min     0
```

```
Fermion_omega_max     0
Fermion_EFermi        1e4

outfile              Fermion_N2.out
```

`Fermion_*` indicates that what comes after is a parameter for the Fermionic environment specified by the above Hamiltonian. `Fermion_N_modes` tells the code to use 2 Fermionic states as environment. The coupling strength is determined by `Fermion_g` and the energies are equidistantly sampled from `Fermion_omega_min` to `Fermion_omega_max` (in inverse picoseconds; there also exist the alternative `Fermion_E_min` and `Fermion_E_max` to specify the band width in units of meV). Here, both limits are set to zero, so that both environment modes are resonant to the TLS transition. By setting `Fermion_EFermi 1e4` the Fermi level is set to such a high value that all environment states are initially occupied. There is also the parameter `Fermion_temperature` to specify the temperature (in units of Kelvin) of the Fermi distribution. If not specified, the global `temperature` parameter will be used, whose default is 0 K.

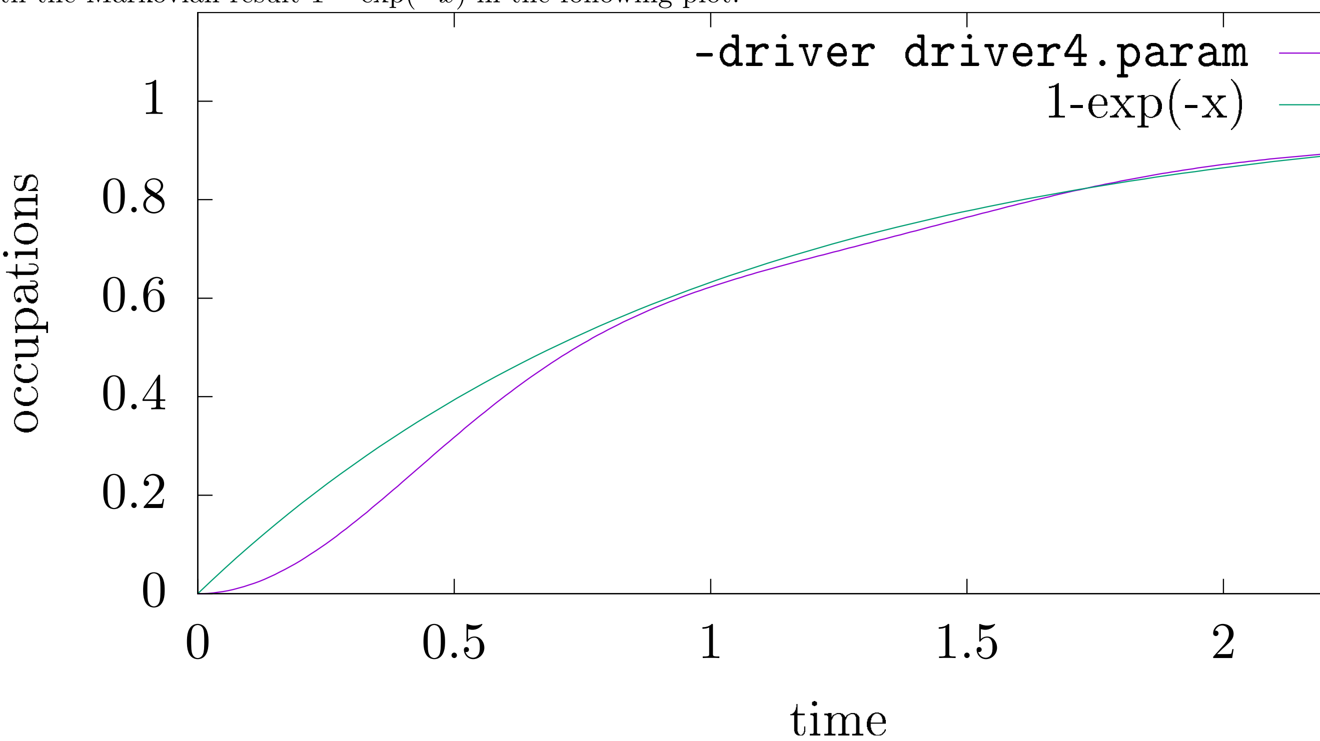
Here, the initial state of the system is empty. Therefore, electrons will start to move from the Fermionic environment to the system. The dynamics is show below and it is discussed in the ACE article.



Typically, the environments of open quantum system are assumed to form a continuum. In ACE, we simply discretize the continuum. For the case of metallic leads, it turns out that using $N_E = 10$ modes is already not too bad. Consider the driver file `Fermion.param`:

```
te                    2.5
dt                    1e-2
threshold             1e-7

Fermion_N_modes        10
Fermion_rate            1
Fermion_omega_min      -5
Fermion_omega_max       5
Fermion_EFermi          1e4

outfile               Fermion_N10.out
```

Here, instead of **`Fermion_g`**, we use **`Fermion_rate`** to specify the rate that we would expect in the Markov limit. Then, the coupling constant is calculated internally by solving the Fermi's golden rule expression for g. The respective dynamics is compared with the Markovian result $1 - \exp(-x)$ in the following plot:



Increasing N_E even further to about 100 while keeping the same density of states (i.e. increasing the band width simultaneously) will produce a behaviour very close to the Markovian results.

5.2 Bosonic environments

The class of `Boson_*` environments covers environment Hamiltonians of the form

$$H_E = \sum_{\mathbf{k}} \Big[\hbar\omega_{\mathbf{k}} a^\dagger_{\mathbf{k}} a_{\mathbf{k}} + \hbar g_{\mathbf{k}} \big(a^\dagger_{\mathbf{k}} \hat{O}_{sys} + a_{\mathbf{k}} \hat{O}^\dagger_{sys} \big) \Big]. \tag{10}$$

In the case $\hat{O}_{sys} = |1 >< 1|_2$, this leads to the independent boson model, which is often used to model the effects of vibrational baths (phonons). If instead $\hat{O}_{sys} = |0 >< 1|_2$ is used, the Hamiltonian describes Jaynes-Cummings interactions, e.g., a quantum emitter coupled to photon modes. This name of the parameter for this operator is `Boson_SysOp`.

The command line arguments are similar to that for the Fermionic case, only that `Fermion_*` is replaced by `Boson_*`. The main differences to the Fermionic case are that initial thermal states now follow Bose statistics and that the number of excitations per mode is, in principle, unbounded. Here, we truncate the Boson Hilbert space per mode and only account for the M lowest states, i.e. $M-1$ excitations. This information if provided to the code by the parameter `Boson_M`, whose default value is $M = 2$.

As a first example, consider the radiative decay of an initially excited two-level quantum emitter. The corresponding parameter file `radiative_decay.param` is

```
te                  2.5
dt                  1e-1
threshold           1e-5

Boson_N_modes       20
Boson_SysOp         {|0><1|_2}
Boson_rate           1
Boson_omega_min    -10
Boson_omega_max     10
Boson_temperature    0

initial             {|1><1|_2}

outfile             radiative_decay.out
```

The resulting dynamics resembles the Markovian result with excited state occupations $\langle(|1\rangle\langle 1|)\rangle = \exp(-t)$. (Again, an environment with larger band width would lead to a more Markovian behaviour, but has to be discretized with more modes and therefore takes longer to calculate.).

One caveat for calculations at finite temperatures: The Jaynes-Cummings Hamiltonian acts in the rotating frame. This means that the physical energy of a Bosonic excitation is actually $(\hbar\omega + E_{shift})n$, where E_{shift} is the energy shift corresponding to the frequency of the rotating frame. Hence, negative values of ω are physically allowed if their modulus is smaller than E_{shift}. For initial thermal states, this value can be provided by

Boson_E_shift_init or Boson_omega_shift_init. The latter is internally multiplied by $\hbar$.

For structured baths where every environment mode is coupled to the system with a different strength, i.e., g_k is not constant as a function of k, there are different ways to pass the values for $E_k = \hbar\omega_k$ and g_k to the code. One way is to compile a file with two columns, the first listing the values of E_k (or ω_k) and the second listing the corresponding values of g_k. Please make sure the number of lines matches Boson_N_modes. The name of this file can be passed to the code by the argument Boson_E_g_from_table (or Boson_omega_g_from_table). For example, if we create the file "N20.tab" with the 20 lines

```
-9.5   {1/sqrt(2*pi)}
-8.5   {1/sqrt(2*pi)}
-7.5   {1/sqrt(2*pi)}
-6.5   {1/sqrt(2*pi)}
...
 9.5   {1/sqrt(2*pi)}
```

the following driver file

```
te                    2.5
dt                    1e-1
threshold             1e-5

Boson_N_modes         20
Boson_SysOp           {|0><1|_2}
Boson_omega_g_from_table   N20.tab
Boson_temperature      0

initial               {|1><1|_2}

outfile               radiative_decay_tab.out
```

exactly reproduces the results in "radiative_decay.out".

Another way, which is highly recommended, is to use instead a spectral density defined by

$$J(\omega) = \sum_k g_k^2 \delta(\omega - \omega_k) \tag{11}$$

which is assumed to be a continuous function of ω in the limit of infinitely fine discretization. The spectral density has a more intuitive interpretation. For example, in the case of radiative decay, the radiative decay rate in the Markov limit of a system driven with frequency ω is $2\pi J(\omega)$, a finding which we reproduce numerically next. In contrast, to reproduce a given rate, the values of g_k have to be modified when the discretization changes.

A spectral density can be provided to the ACE code in the form of a file that contains two columns corresponding to a set of sample points ω_i and $J(\omega_i)$. The sample points

ω_i are not related to the discretization used for defining the set of environment modes. Instead, the frequency domain from Boson_omega_min to Boson_omega_max is discretized into Boson_N_modes intervals and the corresponding values for g_k are obtained by linearly interpolating between the closest sample points to ω_k in the spectral density file. This way, the spectral density file has to be generated only once and can be reused for calculations with different mode discretizations. In the following example, we reproduce the above result for radiative decay, now using a flat spectral density. To this end, generate the file Jflat.J with the following two lines

```
-100. 1.
 100. 1.
```

Therefore, all interpolated values of $J(\omega)$ will be 1. We then use the parameter file

```
te                   2.5
dt                   1e-1
threshold            1e-5

Boson_N_modes        20
Boson_SysOp          {|0><1|_2}
Boson_J_from_file     Jflat.J
Boson_J_scale         {1/(2*pi)}
Boson_omega_min     -10
Boson_omega_max      10
Boson_temperature     0

initial              {|1><1|_2}

outfile              Jflat.out
```

The parameter Boson_J_scale rescales the spectral density by a global factor for all sample points, in this case $1/(2\pi)$, so the expected Markovian rate will be $2\pi J(\omega) = 2\pi(1/(2\pi)) = 1$. Please check that this approach reproduces exactly the result for radiative decay discussed above.

5.3 Predefined spectral densities

Some commonly used spectral densities are predefined and can be enabled by the command line argument Boson_J_type. For example, one often uses spectral densities of the form

$$J(\omega) = \alpha\omega^s H(\omega), \tag{12}$$

which is characterized by the exponent s distinguishing ohmic $s = 1$ from sub- ($s < 1$) and superohmic ($s > 1$) spectral densities. α sets the strength of the system-bath coupling and $H(\omega)$ is a cut-off function used to make the large-ω limit well defined. This type of spectral density is activated by setting Boson_J_type to ohmic. α and s can be set via Boson_J_alpha and Boson_J_s, respectively. The finite energy range defined by Boson_omega_min and Boson_omega_max already leads to a natural cutoff, but one can also use, e.g., an exponential cutoff $H(\omega) = exp(-\omega/\omega_c)$ by setting Boson_J_cutoff to exp and Boson_J_omega_c to ω_c.

Keep in mind that the generated spectral density can be printed by providing a file name to the parameter Boson_J_print, which can be useful for debugging.

The independent boson model, i.e, a TLS diagonally and linearly coupled to a continuum of harmonic oscillators, is also a good model for the coupling between electronic states in a quantum dot (QD) and longitudinal acoustic phonons. The corresponding Hamiltonian is

$$H_E = \sum_{\mathbf{q}} \Big[\hbar\omega_{\mathbf{q}} b^\dagger_{\mathbf{q}} b_{\mathbf{q}} + \hbar\gamma_{\mathbf{q}} \big(b^\dagger_{\mathbf{q}} + b_{\mathbf{q}} \big) |X\rangle\langle X| \Big], \tag{13}$$

where $b^\dagger_{\mathbf{q}}$ and $b_{\mathbf{q}}$ are creation and annihilation operators for phonons with wave vector $\mathbf{q}$.

Due to the ordered structure of solid state crystals, electron-phonon interactions are well understood and can be derived from microscopic considerations. It turns out that the dominant deformation potential coupling to longitudinal acoustic phonons is superohmic with exponent $s = 3$. For a typical GaAs-based semiconductor quantum dot, a set of parameters that enter the spectral density has been worked out by Krummheuer *et al.* in [Phys. Rev. B 71, 235329 (2005)]. To use them, simply set Boson_J_type to QDPhonon. Try the following parameter file QDPhonon_ACE.param:

```
te                     20
dt                      1e-1
Nintermediate          20

threshold               5e-8
dict_zero               1e-12

add_Hamiltonian              {-1.5*|1><1|_2}
add_Pulse Gauss  7 5 3 0     {|1><0|_2}

Boson_N_modes                      100
Boson_M                              3
Boson_E_max                          5
Boson_J_type                         QDPhonon
Boson_subtract_polaron_shift         true
temperature                          4

outfile                              QDPhonon_ACE.out
```

Two parameters in this file have not been discussed so far, dict_zero and Boson_subtract_polaron_shift. The former enables a trick to increase efficiency based on group decomposition as discussed in the supplementary material of the ACE article on the example of superradiance, which is also related to [Phys. Rev. B 96, 201201(R) (2017)] describing an analogous idea for the iQUAPI method: For certain types of couplings, MPO matrices $\mathcal{Q}^{(\alpha_l,\tilde{\alpha}_l)}_{d_l d_{l-1}}$ are identical for different combinations of $(\alpha_l, \tilde{\alpha}_l)$. In this case, only one representation has to be stored and calculated, which reduces the numerical demands. In the above example with diagonal coupling (`Boson_SysOp` has the default value of `|1><1|_2`), the environment does not induce system transitions, so $\mathcal{Q}^{(\alpha_l,\tilde{\alpha}_l)}_{d_l d_{l-1}} = 0$ for $\alpha_l \neq \tilde{\alpha}_l$. These don't have to be stored, reducing the numerical demands by at least a factor of 4 (2^2 instead of 2^4 combinations of $(\alpha_l, \tilde{\alpha}_l)$). Which matrices are zero and which are identical with matrices for other combinations of system indices can be computed automatically from the microscopic Hamiltonian. The parameter `dict_zero` tells the code what can be considered as practically zero or practically identical for this purpose. If a positive value is given, automatic detection of groups with identical couplings is enabled.

Setting `Boson_subtract_polaron_shift` to `true` tells the code to subtract polaron shift from the system energy levels: For the independent boson model, it is well known that the interaction with the bath renormalizes the energies of the system by the polaron shift $\Delta E_p = -\sum_{\mathbf{q}}(\gamma^2_{\mathbf{q}})/(\omega_{\mathbf{q}}) = -\int\limits_0^\infty d\omega\, J(\omega)/d\omega$. As this shift is present irrespective of the state of the system or the environment, experimental determination of the energy levels of the TLS usually reveals the polaron shifted values. It is therefore convenient to subtract the polaron shift, redefine the energy levels, and avoid dealing with renormalization effects explicitly when comparing calculations with and without phonons.

5.4 Related computational methods

Finally, as an alternative to ACE, the process tensor for Gaussian baths can be calculated using expressions where the bath is already integrated out [cf. Jørgensen and Pollock Phys. Rev. Lett. 123, 240602 (2019)]. This method is usually much more efficient and does not require discretization or truncation of the phonon Hilbert spaces (Recall: The advantage of ACE is its generality, while the latter method only works for Gaussian baths.) To use this method instead for phonon simulations with our standard QD phonon spectral density, one only needs to set `use_Gaussian` to `true`. For example, we can re-use the parameter file from the last example and run:

```
> ACE QDPhonon_ACE.param -use_Gaussian true -outfile QDPhonon_Gaussian.out
```

With the ACE code, we also provide binaries for the iterative methods iQUAPI and TEMPO. If they are compiled, you can also try

```
> TEMPO QDPhonon_ACE.param -outfile QDPhonon_TEMPO.out
```

or

```
> iQUAPI QDPhonon_ACE.param -n_max 12 -dt 0.2 -outfile QDPhonon_iQUAPI.out
```

Note that iQUAPI performs no tensor network compression but instead works by memory truncation, where only a fixed maximal number `n_max` of past time steps are accounted for. Because the memory demands scale exponentially with `n_max`, only relatively small values of `n_max` can be used in practice. To cover the memory time of the environment the time step width `dt` should be choosen as large as possible.

5.5 Reading, writing, and combining process tensors

A PT generated by the ACE cod ecan be written into a binary file with a name specified by the `write_PT` parameter. Such a PT can then be read for another simulation using the `read_PT`. Furthermore, multiple PTs can be loaded and used for the propagation by the `multi_PT` argument. The difference between the use of `read_PT` and `multi_PT` is that when new baths are specified, e.g., by setting `Boson_N_modes` or `Fermion_N_modes` to nonzero values or using an `add_single_mode` line, these new baths will be incorporated into a PT specified by `read_PT`, while the PTs defined by `multi_PT` are only loaded for the propagation of a concrete simulation.

6 Concluding remarks

Further developments of ACE, the method as well as the code, are ongoing projects. Some implemented features are not described in the documentation yet, but will be added in the future.