



## Quantum Dynamics E-CAM modules V

E-CAM Deliverable 3.6

Deliverable Type: Other

Delivered in March, 2021



E-CAM

The European Centre of Excellence for  
Software, Training and Consultancy  
in Simulation and Modelling



Funded by the European Union under grant agreement 676531

### Project and Deliverable Information

Project Title	E-CAM: An e-infrastructure for software, training and discussion in simulation and modelling
Project Ref.	Grant Agreement 676531
Project Website	<a href="https://www.e-cam2020.eu">https://www.e-cam2020.eu</a>
EC Project Officer	Juan Pelegrín
Deliverable ID	D3.6
Deliverable Nature	Other
Dissemination Level	Public
Contractual Date of Delivery	Project Month 62(30 <sup>th</sup> November, 2020)
Actual Date of Delivery	19 <sup>th</sup> March, 2021
Deliverable Description	Final software modules delivered to the E-CAM library in the area of Quantum Dynamics based on users requests and their documentation.

### Document Control Information

Document	Title:	Quantum Dynamics E-CAM modules V
	ID:	D3.6
	Version:	As of 19 <sup>th</sup> March, 2021
	Status:	Accepted by WP leader
	Available at:	<a href="https://www.e-cam2020.eu/deliverables">https://www.e-cam2020.eu/deliverables</a>
Review	Document history:	<a href="#">Internal Project Management Link</a>
Review	Review Status:	Reviewed
Authorship	Written by:	Ahai Chen(CEA-Saclay)
	Contributors:	F. Agostini (Université Paris-Saclay), S. Bonella (EPFL), A. Chen (CEA-Saclay) S. Gómez (University of Vienna), M. Heindl (University of Vienna), D. Lauergrnat (Université Paris-Saclay), K. Parsons (Dalhousie University) C. Sanz (Autonomous University of Madrid), T. Tran (UCL), G. Worth (UCL), Ana Catarina Mendonça (EPFL)
	Reviewed by:	Sara Bonella (EPFL)
	Approved by:	Sara Bonella (EPFL)

### Document Keywords

Keywords:	E-CAM, Quantum Dynamics, Module, Potential Library, Surface Hopping, Exact Factorization, Interface, Smolyak method, CECAM
-----------	--

19<sup>th</sup> March, 2021

**Disclaimer:** This deliverable has been prepared by the responsible Work Package of the Project in accordance with the Consortium Agreement and the Grant Agreement. It solely reflects the opinion of the parties to such agreements on a collective basis in the context of the Project and to the extent foreseen in such agreements.

**Copyright notices:** This deliverable was co-ordinated by Ahai Chen<sup>1</sup> (CEA-Saclay) on behalf of the E-CAM consortium with contributions from F. Agostini (Université Paris-Saclay), S. Bonella (EPFL), A. Chen (CEA-Saclay) S. Gómez (University of Vienna), M. Heindl (University of Vienna), D. Lauergrnat (Université Paris-Saclay), K. Parsons (Dalhousie University) C. Sanz (Autonomous University of Madrid), T. Tran (UCL), G. Worth (UCL), Ana Catarina Mendonça (EPFL)

This work is licensed under the Creative Commons Attribution 4.0 International License. To view a copy of this license, visit <http://creativecommons.org/licenses/by/4.0>.



<sup>1</sup>[ahai.chen@cea.fr](mailto:ahai.chen@cea.fr)

# Contents

<b>Executive Summary</b>	<b>1</b>
<b>1 Introduction</b>	<b>2</b>
1.1 Overall scope of the module set . . . . .	2
1.2 General applications and possible exploitation of the codes . . . . .	3
<b>2 Modules</b>	<b>4</b>
2.1 G-CTMQC . . . . .	4
2.1.1 Scientific context . . . . .	4
2.1.2 Practical application and exploitation of the code . . . . .	4
2.2 FBTS-MPI . . . . .	4
2.2.1 Practical application and exploitation of the code . . . . .	5
2.3 Quantics DD-vMCG MPI/OMP . . . . .	5
2.3.1 Practical application and exploitation . . . . .	5
2.4 SHARC-gym . . . . .	5
2.4.1 Practical application and exploitation of the code . . . . .	6
2.5 ElVibRot-TID-MPI . . . . .	6
2.5.1 Practical application and exploitation of the code . . . . .	7
2.6 ElVibRot-TD-MPI . . . . .	7
2.6.1 Practical application and exploitation of the code . . . . .	7
<b>3 Overall impact of the results achieved within the Work-package</b>	<b>8</b>
3.1 Overview of the results achieved so far . . . . .	8
3.2 Dissemination and exploitation . . . . .	8
3.3 Industrial impact . . . . .	9
3.4 Training . . . . .	9
3.5 Societal impact . . . . .	10
<b>4 Conclusion and Outlook</b>	<b>11</b>
<b>References</b>	<b>12</b>

## Executive Summary

In this report for Deliverable 3.6 of E-CAM, 6 software modules in quantum dynamics are presented. All modules stem from the activities initiated during the *SAW* held at Lyon (France) in June 2019 and the workshop *ESDW: Quantum Dynamics*, held at Durham University (UK) in July 2019. The modules originate from the input of E-CAM's academic user base. They have been developed by members of the project (S. Bonella EPF-L), established collaborators (G. Worth University College London, S. Gomez University of Vienna, C. Sanz University of Madrid, D. Lauvergnat Paris-Saclay University, F. Agostini Paris-Saclay University), E-CAM post-doc (A. Chen CEA-Saclay) and new contributors to the E-CAM repository (M. Heindl University of Vienna, K. Parsons Dalhousie University, T. Tran University College London). The presence of new contributors indicates the interest of the community in our efforts. Furthermore, the contributors to modules in WP3 continue to be at different stages of their careers (in particular, K. Parsons, T. Tran, and M. Heindl are PhD students) highlighting the training value of our activities.

Following the order of presentation, the 6 modules are named: *G-CTMQC*, *FBTS-MPI*, *Quantics DD-vMCG MPI/OMP*, *SHARC-gym*, *ElVibRot-TID-MPI*, and *ElVibRot-TD-MPI*. In this report, a short description is written for each module, followed by a link to the respective Merge-Request document on the GitLab service of E-CAM. These merge requests contain detailed information about the code development, testing and documentation of the modules.

As this deliverable is the last one of its series, a section on the overall impact of the results achieved within the Workpackage Quantum Dynamics was also included, with an overview of the results achieved so far, how these have been disseminated in scientific publications, the industrial impact of the software developed, the training given in the WP which has led to many of the software outputs, and the societal impact of the WP.

# 1 Introduction

Work Package 3 (Work-package (WP)3) of E-CAM provides a means for academic and industrial users of addressing computational questions that involve quantum dynamics, in particular, by developing scalable, open source software with verified quality standards, appropriate documentation and testing. E-CAM's activities in this area can be divided into two, connected, main pillars: the production of modules solicited by our user base (academic and industrial) and the creation of an environment to foster the transition from in-house software to high performing and reliable community packages with long-term sustainability. Within the scope of this deliverable and based on the plan of activities outlined in deliverable D3.2: Identification/Selection of E-CAM Quantum Dynamics Codes for Development [1], we have:

- Developed modules implementing different Quantum-Classical dynamics methods, with particular reference to nonadiabatic phenomena;
- Constructed interfaces between quantum dynamics codes within WP3 and external electronic structure codes to obtain potentials and forces necessary for dynamics calculations;
- Expanded the functionalities and the efficiency of E-CAM WP3 flagship codes, PaPIM and QUANTICS.
- Developed a parallel version of a third reference package for E-CAM WP3, ElVibRot

To describe each module, a module documentation repository is created in an [E-CAM GitLab repository](#) which automatically updates documentation for the [E-CAM Quantum Dynamics Module Documentation](#) on [Read the Docs](#). To submit a module, a developer can fork the GitLab repository and create his own feature branch in which s/he documents the latest progress about module development, source code link/patch file, new feature commit, etc. Developers can submit a merge request of their branch into the E-CAM master branch via an established Merge-Request procedure. The E-CAM software manager and developers then run the module through the project's acceptance criteria (coding style and structure, source code documentation and module testing). The software manager then accepts the merge or rejects it providing at the same time feedback and, when necessary, assistance to overcome the reasons for the rejection. The documentation will only appear as part of the [Work Package 3 Software Modules](#) on [ReadTheDocs.org](#) once the Merge-Request is accepted.

In section 2, a short description is provided for each reported module, followed by a link to the Merge-Request on the GitLab service of E-CAM, where more detailed information about code development, testing and documentation can be found. A section dedicated to the practical application and exploitation of the code is also presented for each module.

## 1.1 Overall scope of the module set

This module set further expands the scope of E-CAM's production of software in the area of quantum dynamics via two new main venues identified in the overall development plan described in deliverable D3.2.[1]. The first set of modules (G-CTMQC, FBTS-MPI) implements in the E-CAM library approximate methods in the area of mixed quantum-classical dynamics with particular focus on non-adiabatic processes; the second set of modules (Quantics DD-vMCG MPI/OMP, SHARC-gym, ElVibRot-TID-MPI, ElVibRot-TD-MPI) makes further extension and improvement of QUANTICS (one of E-CAM's main codes in WP3) and ElVibRot (another code identified as a potential E-CAM flagship) and continues to provide useful tools for an efficient and easy accessibility crossing the interface between quantum dynamics simulations and electronic structure calculations.

As mentioned in the previous deliverable, non-adiabatic processes (i.e. phenomena in which nuclear motion, external probes, or environmental effects cause transitions between the quantum electronic states of a system) are becoming increasingly relevant in areas ranging from the manipulation of materials via photochemical or photophysical processes (e.g. conversion of light to electricity in solar cells, mimicking photosynthetic processes in new devices, reactions in fuel cells), to health (e.g. in studying the radiation induced damage to DNA that eventually lead to skin cancer), or in optimizing and guiding chemical reactions towards desired products via laser pulses. Brute force methods for these problems, however, are numerically too expensive to be of practical use, leading to the development of several approximate methods as described in detail in D3.2: Identification/Selection of E-CAM Quantum Dynamics Codes for Development [1] and in the scientific report from the Lyon E-CAM State-of-the-art workshop "Recent developments in quantum dynamics" (June 2019) that is available [here](#). The modules presented here, based on D3.2[1] and D3.3[2], implement state-of-the-art algorithms in the domain and extend, by recruiting new codes and methods, the scope of the E-CAM repository.

G-CTMQC(see section 2.1) provides a tool to perform excited-state molecular dynamics simulations with various trajectory-based algorithms, including non-adiabatic coupling and spin-orbit coupling. FBTS-MPI implements the Forward-Backward Trajectory Solution to the quantum-classical Liouville equation developed by Hsieh and Kapral [3, 4] in the study of non-adiabatic quantum dynamics.

The second set of modules provides further improvements and extensions to two E-CAM codes, [QUANTICS](#) and [ELVibRot](#). In particular, [Quantics DD-vMCG MPI/OMP](#) provides an improved version of the parallel code for Direct Dynamics variational multi-configuration Gaussian (DD-vMCG) method and the Quantum-Ehrenfest variant in Quantics. [SHARC-gym](#) builds a bridge between surface hopping and more accurate methods such as multi-configurational time dependent Hartree (MCTDH) and variational multi-configurational gaussian (vMCG) by exploiting both types of methods to overcome the shortcomings of the other in a hybrid approach. [ELVibRot-TID-MPI](#) and [ELVibRot-TD-MPI](#) provides a MPI implementation for [ELVibRot](#) package to support parallel time-independent quantum simulation or wavepackage propagation of general molecular system.

## 1.2 General applications and possible exploitation of the codes

The modules presented are aimed principally at the academic user base in E-CAM. This is in itself an important target because, due to the novelty and complexity of the area of nonadiabatic dynamics, no well established community software exists and E-CAM is playing an important role in creating a reference library and a set of well maintained training and research tools for the growing community of users in this area. Furthermore, the codes developed have the potential to enable applications in domains of societal impact and technological interest.

## 2 Modules

In this section, a short description is written for each module, followed by a link to the Merge-Request on [GitLab service of E-CAM](#), which shows detailed information about code development, testing and documentation.

### 2.1 G-CTMQC

G-CTMQC is a module to perform excited-state molecular dynamics simulations with various trajectory-based algorithms, including nonadiabatic coupling and spin-orbit coupling.

Nuclear dynamics can be performed based on the quantum-classical algorithm derived from the exact factorization of the electron-nuclear wavefunction [5], dubbed CT-MQC [6]. Recently, the extension of the exact-factorization theory has been proposed to include spin-orbit coupling [7]. Therefore, the “generalized” algorithm is now able to treat (i) standard nonadiabatic situations, where spin-allowed electronic transitions among states with the same spin multiplicity are mediated by the coupling to nuclear motion, and (ii) spin-orbit interactions, where spin-forbidden electronic transitions among states of different spin multiplicity are induced by the spin-orbit coupling.

Electronic evolution is carried out in the adiabatic basis for standard nonadiabatic problems. In the case of spin-orbit interactions, G-CTMQC offers the option to use the spin-diabatic or the spin-adiabatic representations. Information about electronic-structure properties, i.e. energies, gradients and couplings, is calculated and read on-the-fly at the positions of the trajectories at each time step based on the ModelLib library [8] of potentials (which G-CTMQC is interfaced to).

In addition, the code offers the possibility of performing calculations with the trajectory surface hopping algorithm [9] and the Ehrenfest approach [10]. Concerning the trajectory surface hopping method, the fewest switches scheme is implemented, along with the energy decoherence corrections to fix the overcoherence issue of surface hopping [11]. For surface hopping and Ehrenfest, only nonadiabatic couplings are currently implemented.

#### 2.1.1 Scientific context

Studies in the domain of photochemistry and photophysics strongly rely on simulation methods able to describe strong coupling between electronic and nuclear motion on the femtosecond time scale, usually called nonadiabatic coupling. Simulations give access to microscopic information in terms of molecular structures, electronic populations, vibrational energies that can be easily compared to experiments, for instance in the domains of time-resolved spectroscopy or 2D spectroscopy. In addition to this, molecular dynamics simulations allow to follow in real time the evolution of molecular systems, thus providing support to interpret and even predict the outcome of experiments.

Photochemical and photophysical reactions are ubiquitous in nature, from photosynthesis to vision, and are more and more exploited for technological advances, as for the photo-current production in organic photovoltaic devices. In addition, it is becoming clear the importance to consider spin-orbit coupling even in those systems composed of light elements, such as oxygen and carbon, to be able to describe processes such as intersystem crossings in organic light-emitting diodes.

#### 2.1.2 Practical application and exploitation of the code

G-CTMQC module provides numerical tools to perform simulations of internal conversion (spin-allowed) and intersystem crossing (spin-forbidden) phenomena underlying photochemical and photophysical reactions. G-CTMQC gives the user the flexibility of employing different approaches and, thus, various approximation schemes, to achieve dynamical information as accurate as possible, as well as ample flexibility in the choice of systems to be studied, thanks to the interface of G-CTMQC with ModelLib (E-CAM module).

G-CTMQC module provides the basis for future computational developments planned in the framework of the recently-funded project Q-DeLight by the French National Agency for Research (ANR) coordinated by Federica Agostini (2021-2024). Furthermore, it has been used for several scientific publications [12, 13, 14].

Merge Request	<a href="#">Merge-Request of G-CTMQC module.</a>
Direct Documentation Link	<a href="#">readme.rst of G-CTMQC module.</a>

### 2.2 FBTS-MPI

The FBTS-MPI module implements the Forward-Backward Trajectory Solution (FBTS) to the quantum-classical Liouville equation [15] developed by Hsieh and Kapral in the study of non-adiabatic quantum dynamics [3, 4]. In this

approximate quantum dynamics method a many-body system is partitioned into a quantum subsystem that is coupled to a classical-like environment, wherein both the subsystem and environment degrees of freedom are transformed into a continuous phase space representation. This allows for the FBTS-MPI module to evolve an ensemble of Monte Carlo sampled trajectories through time according to classical-like equations of motion. The purpose of this module is the construction of time-dependent observables that can be used in the study of the dynamics of these systems.

### 2.2.1 Practical application and exploitation of the code

The particular system that this FBTS-MPI module has been built for is in the study of excitation energy transfer in biological light harvesting systems, so-called protein-pigment complexes, through the use of the Frenkel exciton model. In this model the quantum subsystem of interest is the electronic excited states of the pigment molecules, and the surrounding vibrational environment is represented as a collection of harmonic oscillators. The initial application for this module is in examining the mechanisms of exciton transport, which can be studied through the time-dependent exciton site populations for a given light-harvesting complex. The approximate nature of this dynamics method combined with the parallelization of the trajectory ensemble allows one to model exciton transport in large systems with many pigments that would otherwise be prohibitively expensive to simulate.

Merge Request	<a href="#">Merge-Request of FBTS-MPI module.</a>
Direct Documentation Link	<a href="#">readme.rst of FBTS-MPI module.</a>

## 2.3 Quantics DD-vMCG MPI/OMP

The Quantics DD MPI/OMP module is an improved version of the parallel code for DD-vMCG method [16, 17] and the Quantum-Ehrenfest variant which are included in the Quantics package program [18].

The Direct Dynamics is a method to solve the time-dependent Schrödinger equation using a variational solution and where the potential energy surface is evaluated *on-the-fly*. This method allows the investigation of quantum effects on the chemical reactivity of a molecule. The DD-vMCG code of Quantics is interfaced to many electronic structure programs and thus, dynamics can be performed with a wide variety of methods as long as they can provide the energy, gradients (and Hessians).

### 2.3.1 Practical application and exploitation

The Quantics package grew out of the Heidelberg MCTDH Package [19].

The module is focused on the DD-vMCG method which allows the study of nonadiabatic dynamics of molecules to investigate for example the photodissociation of 3-pyrroline,[20] or energy dissipation in thymine.[21]

In the DD-vMCG method, at every step of the Direct dynamics propagation, the energies, gradients, and Hessians are evaluated at the center of each gaussian wavepacket by calling an external program. One of the challenge of Direct Dynamics is the cost of computation for the evaluation of the potential energy surfaces. The electronic structure calculations are performed for each gaussian wavepacket individually and thus, parallelizing the call to the electronic structure program greatly decreases the time of computation between each nuclear step.

The implementation of the MPI parallelization is available in [Quantics release 1.3](#).

Merge Request	<a href="#">Merge-Request of Quantics DD-vMCG MPI/OMP module.</a>
Direct Documentation Link	<a href="#">readme.rst of Quantics DD-vMCG MPI/OMP module.</a>

## 2.4 SHARC-gym

This module aims at building a bridge between surface hopping (SH)[22] and more accurate methods (summarized with the term QUANTUM in the following) like MCTDH[23] and vMCG[24] by exploiting both types of methods to overcome the shortcomings of the other in a hybrid approach called the SHARC-gym[25].

In the computational simulation of molecular movements and reactions, various degrees of simplification have been introduced. From exact quantum dynamics available only to model a few degrees of freedom up to huge coarse-grained simulations capable to model whole proteins different levels of sophistication are available. Exact quantum dynamics and methods that will converge to the exact result are capable to shed insight into the most intricate of mechanisms at the heart of processes like photosynthesis. Unfortunately, the use of these methods is hampered by the unfavorable scaling of the simulation time with the size of the investigated system, limiting those approaches to a few dozen degrees of freedom. During the last decades, surface hopping has risen to be one of the most popular approaches for the simulation of events that involve more than a single electronic state and more than 10 atoms. This popularity is due to the ease of implementation of an SH algorithm and the possibility to plug in properties

calculated using any of the most popular quantum chemistry packages. However, while the foundations of SH are easy to grasp, the *ad hoc* nature of SH means that there never is any guarantee that the simulated dynamics for a given system resembles results obtained via more elaborate methods that do not suffer such crude approximations. Many of the shortcomings of SH have been highlighted in the scientific literature and remedies to overcome those have been proposed. This means that a whole range of various additional parameters and flavours of SH do exist at the present state that are combined or used exclusively at the will of the user, hoping that these corrections will result in more accurate modeling of the problem at hand.

The SHARC-gym allows the user to overcome this uncertainty by combining SH and QUANTUM methods in a hybrid fashion. The method follows an iterative procedure which is briefly stated here:

1) *Hamiltonian loop*: The aim of this loop is to select the most important degrees of freedom using SH so that a stripped-down Hamiltonian can be used in QUANTUM dynamics. For this, a full-dimensional SH dynamics is conducted which serves as a reference throughout this loop. From this full-D SH reference, the degrees of freedom (molecular vibrations, movement or even electronic states) that drive the observed dynamics can be determined. Using these essential degrees of freedom, a new model with reduced dimensionality is constructed and a new SH simulation calculated. If this new simulation still contains the most important features of the dynamics, even more degrees of freedom can be cut from the Hamiltonian and the SH dynamics is repeated. Once too many modes have been stripped away and the results diverge from the full-D SH reference, this process is stopped and the Hamiltonian that was used before this last dynamics is used in the subsequent *Parameter loop*.

2) *Parameter loop*: In this loop, the reduced Hamiltonian is used in a QUANTUM simulation which serves as a QUANTUM reference throughout the loop. Now that a QUANTUM reference in this reduced Hamiltonian is available, the plethora of parameters available in SH can be validated for this system. If the initially used set of SH parameters was found to perform well, then the SHARC-gym is finished, resulting in a QUANTUM-validated set of parameters for the full-D SH dynamics and a reduced Hamiltonian that captures the essential dynamics of the much bigger system. If the best set of SH parameters diverges from the set that has been used to determine the reduced Hamiltonian, this new set of parameters has to be used again in the *Hamiltonian loop* and the process has to be repeated as a whole until the best agreement is found.

#### 2.4.1 Practical application and exploitation of the code

The hybrid approach of the SHARC-gym enables the use of more accurate QUANTUM methods on a subset of degrees of freedom of larger systems that - as a whole - cannot be treated using a QUANTUM method. This selection of important degrees of freedom is based solely on other dynamics result, eliminating the bias of selecting a set of reactive coordinates beforehand. The SH dynamics benefit from a validation of the chosen parameters against the QUANTUM reference. Furthermore, the SHARC-gym provides a huge amount of possible test systems to test the shortcomings of different parameters of SH or even SH as a whole as the SHARC-gym may result in a QUANTUM reference which disagree with all the different flavours of SH.

The current implementation of the SHARC-gym uses the SH code SHARC[26, 27] and the set of QUANTUM methods implemented in QUANTICS[28].

Merge Request	<a href="#">Merge-Request of SHARC-gym module.</a>
Direct Documentation Link	<a href="#">readme.rst of SHARC-gym module.</a>

## 2.5 EIVibRot-TID-MPI

The EIVibRot time-independent MPI (EIVibRot-TID-MPI) module is a parallelized time-independent quantum simulation program. The Davidson algorithm is the main method employed for getting the Eigen levels of the Hamiltonian. This module is a part of the EIVibRot [29] package designed for general quantum dynamics simulation using curvilinear coordinates. The code has no built-in limitation in terms of the number of degrees of freedom. It applied a numerical but exact kinetic energy operator with Tnum [30], which enables much flexibility in the choice of the curvilinear coordinates. To avoid the conventional direct-product basis sets and grids, the Smolyak algorithm [31] is employed to make possible the simulation of larger systems.

The core of the quantum simulation lies in solving the Schrödinger equation with the Hamiltonian of the considered system. The principle fence of the simulation comes from the exponential growth of computational demand with the increasing of the degrees of freedom of the system, the curse of dimensionality. It prompts numbers of algorithms in the past decades to deal with this difficulty. The Smolyak algorithm, proposed by Smolyak in 1963 [31], provides a powerful method to deal with high-dimensional problems. By introducing the Smolyak algorithm in this module, the wavefunction is expanded as a weighted sum of small Smolyak wavefunction contributions, thus significantly reducing the computational demand of the simulation. Taking advantage of the structure of the newly transformed wavefunction, the MPI can be well implemented. As a result, the simulation can be performed with high accuracy,

and in the meantime, impressive parallel efficiency. The code is designed to work on different levels of clusters. The module provides three MPI schemes to adapt the simulation of different kinds of systems and working machines. The default setting will automatically choose the scheme according to the balance of resource consumed and the parallelization efficiency.

### 2.5.1 Practical application and exploitation of the code

This module is intended to provide a parallel program for the quantum simulation of general molecular systems. Typically, it could be used to calculate the vibrational levels of molecular systems. The general capability of the simulation could be up to tens of degrees of freedom. The code has been applied for the simulation of Malondialdehyde ( $C_3H_4O_2$ ), which is of 21 degrees of freedom. The parallelization of the code enables the simulation of larger systems. Furthermore, the flexibility of Smolyak sparse grids promises the further interesting application of the code. For instance, the module could be applied for the calculation of floppy molecular systems, e.g. the clathrate hydrogen, up to hundreds of degrees of freedom with a system-bath separation scheme in current framework.

Merge Request	<a href="#">Merge-Request of ElVibRot-TID-MPI module.</a>
Direct Documentation Link	<a href="#">readme.rst of ElVibRot-TID-MPI module.</a>

## 2.6 ElVibRot-TD-MPI

The ElVibRot time-dependent MPI (ElVibRot-TD-MPI) module is a parallelized time-dependent quantum simulation program. It is a part of the ElVibRot [29] package designed for general quantum dynamics simulation using curvilinear coordinates. There is no built-in limitation on the degrees of freedom for the target system. The code employed a numerical but exact kinetic energy operator with Tnum [30]. The Smolyak algorithm [31] is applied to avoid the direct-product basis sets and grids in the simulation.

The quantum dynamics simulation has provided powerful insight into the underlying mechanism of chemical reactions, laser molecular interaction, etc. The simulation, with the conventional expansion on the direct-product basis, is limited by the exponential growth of computational cost with the increase of degrees of freedom. The MCTDH, a well-known package developed with the aim of generality, expands the wavefunction as a sum of Hartree products with single-particle functions, leading to a very efficient wavepackage propagation. The quantum diffusion Monte Carlo and the Feynman path integral approaches get around the problem by avoiding expanding the wavefunction on a basis set. The variational multi-configuration Gaussian applies on-the-fly quantum chemical calculation of the potential energy to approach the quantum effects in the photochemistry. However, the Smolyak method provides another way to deal with high-dimensional problems, without losing accuracy and universality. The application of Smolyak algorithm enables the simulation of large systems (> 12 degrees of freedom) as the wavefunction is expanded as a weighted sum of small Smolyak wavefunction contributions. The MPI parallelization hinges on this approach. The module is designed to work on different levels of clusters. Three MPI schemes are provided in accord with a series of well-known propagation methods, including the Chebyshev, Runge-Kunta, short iterative Lanczos and Taylor expansion, etc. The three MPI schemes correspond to the simulation with the mode of most efficiency, memory saving, and massive cluster parallelization, respectively. The default setting will automatically choose the scheme according to the balance of resource consumed and the parallelization efficiency.

### 2.6.1 Practical application and exploitation of the code

This module is intended to provide a parallel program for general wavepackage propagation. The general capability of the simulation could be up to tens of degrees of freedom. The propagation time could be up to hundreds of femtoseconds with general computation time, according to the selected propagation method. The code has been applied for the simulation of Pyrazine ( $C_4H_4N_2$ ), which is of 24 degrees of freedom. This module could be a practical tool for general quantum molecular simulation, supporting the further study of molecular dynamics in chemical reactions, ultrafast process, etc.

Merge Request	<a href="#">Merge-Request of ElVibRot-TD-MPI module.</a>
Direct Documentation Link	<a href="#">readme.rst of ElVibRot-TD-MPI module.</a>

## 3 Overall impact of the results achieved within the Work-package

### 3.1 Overview of the results achieved so far

As mentioned in section 1, E-CAM's WP3 Quantum Dynamics "provides a means for academic and industrial users of addressing computational questions that involve quantum dynamics, in particular, by developing scalable, open source software with verified quality standards, appropriate documentation and testing". So far within this WP, 33 software modules have been developed (against a target of 30) which were certified according to the E-CAM style guidelines[32]. The modules developed are:

- **Modules for Quantum computing.** Within the [E-CAM Pilot Project on Quantum Computing](#), in collaboration with IBM Research in Zürich, we developed a toolbox of subroutines and functionalities that implement the Local Control Theory (LCT) used for construction of control pulses for tuning universal logical quantum gates in quantum computers. These modules respond to the needs of the industrial partner, who is developing a universally programmable quantum computer designed on superconducting transmon qubits. The modules developed for this purpose are listed [here](#).
- **Modules that expand the functionalities and the efficiency of E-CAM WP3 flagship codes: PaPIM and QUANTICS.**
  - **PaPIM** is a high performance code for calculation of equilibrated system properties (observables). Some properties can be directly obtained from the distribution function of the system, while properties that depend on the dynamics of the system, such as the structure factor, infrared spectrum or reaction rates, can be obtained from the calculation of appropriate time correlation functions. PaPIM samples either the quantum (Wigner) or classical (Boltzmann) density functions and computes approximate quantum and classical correlation functions. The code is highly parallelized and suitable for use on large HPC machines (see [the code's performance analysis](#)). The code's modular structure enables an easy update/change of any of its modules. Furthermore the coded functionalities can be used independently of each other.

The different modules that make up the PaPIM code are available on our software repository [here](#). Such libraries are rarely available to the community in a Fortran program format. Some of the functionalities within the code are specifically designed for computation of infrared spectra, and serve as a template for the user to implement its own functionalities.
  - **QUANTICS** is a package for molecular Quantum Dynamics simulations that simulates quantum nuclear motion by solving the time-dependent Schrödinger equation. The portfolio of modules developed for QUANTICS within E-CAM is composed by extensions to the programme's functionalities developed at E-CAM's Extended Software Development Workshops (see the full list of modules developed for QUANTICS [here](#)).
- **Additional modules that implement state-of-the-art algorithms in the domain of Quantum Dynamics and extend the scope of the E-CAM repository.** These modules also correspond to users needs but don't belong to one of the categories above, such as the modules based on the general quantum dynamics code [ElVibRot](#), used in the [E-CAM Pilot project on code optimization for exact and linearized quantum dynamics](#) and documented [here](#), or the modules that represent useful tools for an efficient and easy accessible interface between quantum dynamics simulations and electronic structure calculations.

The full portfolio of modules developed under WP3 is accessible from the software library for WP3 at <https://e-cam.readthedocs.io/en/latest/Quantum-Dynamics-Modules/index.html>.

65% of the total number of modules developed under WP3 arise from external contributions (i.e. participants to our ESDWs and collaborations which are not directly supported by the project), and 35% arise from internal contributions (i.e. from Postdoctoral research associate (PDRA)s). These numbers reveal the significant impact E-CAM is having on this community. Due to the novelty and complexity of the area of quantum dynamics (both on the ground electronic state and nonadiabatic), no well established community software exists and E-CAM is playing an important role in creating a reference library and a set of well maintained training and research tools for the growing community of users in this area. Our events in the area of WP3 ( ESDWs and SAWs) help catalyse new developments (see for example ref. [33]) and fostering the transition from in-house codes to reliable, modular, scalable and well documented community packages.

### 3.2 Dissemination and exploitation

5 publications in the area of Quantum Dynamics originated from the project so far, and at least two are work in progress.

1. Adiabatic motion and statistical mechanics via mass-zero constrained dynamics, S. Bonella, A. Coretti, R. Vuilleumier and G. Ciccotti, *Phys. Chem. Chem. Phys.* **2020**, 22, 10775-10785, DOI: <https://doi.org/10.1039/D0CP00163E>. Open access version of the document [here](#).
2. A molecular perspective on Tully models for nonadiabatic dynamics, L. M. Ibele and B. F. E. Curchod, *Phys. Chem. Chem. Phys.* **2020**, 22, 15183, DOI: <https://doi.org/10.1039/D0CP01353F>. Open access version of the document [here](#).
3. Local control theory for supercomputing qubits, M. Mališ, P. KI. Barkoutsos, M. Ganzhorn, S. Filipp, D. J. Egger, S. Bonella and I. Tavernelli, *Phys. Rev. A* **2019**, 99, 052316, <https://doi.org/10.1103/PhysRevA.99.052316>. Open access version of the document [here](#).
4. The Fluctuation-Dissipation Theorem as a Diagnosis and Cure for Zero-Point Energy Leakage in Quantum Thermal Bath Simulations, E. Mangaud, S. Huppert, T. Plé, P. Depondt, S. Bonella, F. Finocchi, *J. Chem. Theory Comput.* **2019**, 15, 2863-2880, DOI: <https://doi.org/10.1021/acs.jctc.8b01164>. Open access version of the document [here](#).
5. Sampling the thermal Wigner density via a generalized Langevin dynamics, T. Plé, S. Huppert, F. Finocchi, P. Depondt, and S. Bonella, *J. Chem. Phys.* **2019**, 151, 114114, DOI: <https://doi.org/10.1063/1.5099246>. Open access version of the document [here](#)

The software produced within this WP was also disseminated via conferences and workshops attended by the members of the WP, via the project website (e.g. in success stories, in the modules of the month category, on the newsletter, etc.) - an overview of our dissemination activities towards promoting the results in this WP is [here](#), and through six deliverables produced during the project lifetime and that are listed [here](#) (under the # 3.x).

Several scientific collaborations have been fostered by the activities in this WP. Collaborations among researchers at the EPFL (Sara Bonella), the Sorbonne University (Simon Huppert), Université Paris-Sud (Federica Agostini), ENS Paris (Rodolphe Vuilleumier), Durham University (Basile Curchod), UCL (Graham Worth), IBM Research Zürich (Ivano Tavernelli), among others, have led to master thesis, PhD thesis and post-doctoral research projects which actively contribute to the outcomes of this WP. Furthermore, the interactions between researchers at these events and the output in the form of new methods and software has been crucial in building up other research projects (see for example the project funded through the Agence Nationale de la Recherche (ANR) which will use as basis for future computational developments the module G-CTMQC documented in section 2.1).

### 3.3 Industrial impact

Industry connection within this WP happened through the pilot project on Quantum Computing in collaboration with a theory and simulation group at IBM (see description and outcomes [here](#)). The aim of this pilot project was to develop a new method and dedicated software for designing control pulses to manipulate qubit systems based on the local control theory (LCT). The method is highly robust and requires only qubit parameters (frequencies, coupling terms) as inputs, and can generate state preparation pulses between any number of qubits in just one evolution of the full system. E-CAM contributed to the theoretical developments in the project, and to the production of software for efficient generation of state preparation pulses.

### 3.4 Training

During the lifetime of the projects we organized four ESDWs in the area of Quantum Dynamics:

- ESDW in Quantum Mechanics and Electronic Structure, 27 June - 8 July 2016, CECAM-FR-MOSER, Maison de la Simulation, Saclay, France, [Event website](#). [Workshop report](#);
- ESDW in Quantum MD, 17-28 July 2017, CECAM-IRL, UCD, Dublin, Ireland, [Event website](#). [Workshop report](#);
- ESDW in Quantum Dynamics, 18-29 June 2018, CECAM-FR-MOSER, Maison de la Simulation, Saclay, France, [Event website](#). [Workshop report](#);
- ESDW in Quantum Dynamics, 8-19 July 2019, Durham University, UK, [Event website](#). [Workshop report](#).

85 people were trained at these events (against a target of 60), on topics focused on the more theoretical aspects of quantum molecular dynamics simulations, and also on best programming practices, performance analysis (Scalasca), benchmarking environment (JUBE), parallel-computing techniques with OpenACC and ScaLAPACK, ANVIL for continuous integration, CUDA for GPU programming, among others.

### 3.5 Societal impact

As technology reaches smaller time and length scales, the quantum properties of matter gains relevance for society and industry. Furthermore, the ubiquitous presence of hydrogen in materials (e.g. impurities in steel) and devices for clean energy (proton based batteries) makes these simulations relevant also in macroscopic devices and at ambient conditions. Finally, the recent interest in the potential revolution of quantum computing – both in terms of simulating devices that could be used as q-bits, and developing quantum algorithms for application in areas ranging from chemistry to cryptography – has opened a whole new set of opportunities for interactions with hardware developers.

Quantum dynamical simulations are then increasingly important in many industrial sectors, including hardware design (e.g. coherence and interference effects for quantum control or design of q-bits), pharmaceuticals (tunneling in enzymatic reactions), energy production or storage (when light is used to induce quantum physical or chemical transformations).

Collaborations in these fields are already active. In the context of E-CAM, new simulation methods and algorithms for quantum computing were developed in collaboration with IBM. Similar collaborations involve participants to this WP.

Surface hopping and multiple spawning are methods of choice to simulate the excited-state dynamics of molecular systems. Applications of these techniques to dyes and emitters have been reported at WP workshops, and brought new insights for the design of molecules in domains such as dyesensitized solar cells (collaborative projects, for example with, Dyesol, Greatcell) or organic light emitting diodes (collaborative projects, for example with, BASE, Novaled). Excited state, and in particular non-adiabatic, dynamics is potentially interesting also for pharmaceutical companies, for example in connection to preventing photo-damage (leading to skin cancer). Applications of quantum dynamics techniques are also central to reveal reaction mechanisms of atmospheric molecules. Models for these reactions currently employ experimental data, but usually require to complement insufficient data with very simplified models, often not accurate enough. Using quantum dynamics to circumvent the problem of missing experimental data would lead to more accurate atmospheric composition models, with a direct societal impact related to the study of chemical reactions involving small molecules on current climate changes.

Finally, it is important to stress that although the relative novelty of the field of quantum dynamics compared to classical molecular dynamics or electronic structure calculations, the field is now approaching sufficient maturity to pursue more actively industrial engagement. To promote this, it is important to create communication channels between academia and industry to disseminate the potential applications of the field, and the role of E-CAM is crucial in this regard.

## 4 Conclusion and Outlook

The report of Deliverable 3.6 of E-CAM describes 6 Software modules in quantum dynamics related to the [E-CAM State-of-the-Art Workshop \(SAW\)](#) held at Lyon (France) in June 2019 and to the workshop [Extended Software Development Workshop \(ESDW\): Quantum Dynamics](#), held at Durham University (UK) in July 2019. The modules include codes for mixed quantum-classical nonadiabatic dynamics, and interfaces between codes of interest in WP3 and electronic structure codes. All the modules have been accepted into the E-CAM software library at [this location](#).

As this deliverable is the last one of its series in E-CAM, a section on the overall impact of the results achieved within the Work-package was also included, with an overview of the results achieved so far, how these have been disseminated in scientific publications, the industrial impact of the software developed, the training given in the WP which has produced many of the software outputs and the societal impact of the WP.

## References

### Acronyms Used

**CECAM** Centre Européen de Calcul Atomique et Moléculaire  
**WP** Work-package  
**MCTDH** multi-configurational time dependent Hartree  
**vMCG** variational multi-configurational gaussian  
**ESDW** Extended Software Development Workshop  
**SAW** E-CAM State-of-the-Art Workshop  
**DD-vMCG** Direct Dynamics variational multi-configuration Gaussian  
**PDRA** Postdoctoral research associate

### URLs referenced

#### Page ii

<https://www.e-cam2020.eu> ... <https://www.e-cam2020.eu>  
<https://www.e-cam2020.eu/deliverables> ... <https://www.e-cam2020.eu/deliverables>  
Internal Project Management Link ... <https://redmine.e-cam2020.eu/issues/146>  
[ahai.chen@cea.fr](mailto:ahai.chen@cea.fr) ... <mailto:ahai.chen@cea.fr>  
<http://creativecommons.org/licenses/by/4.0> ... <http://creativecommons.org/licenses/by/4.0>

#### Page 1

SAW ... <https://www.cecama.org/workshop1774/>  
ESDW: Quantum Dynamics ... <https://www.cecama.org/workshop1697/>

#### Page 2

E-CAM GitLab repository ... <https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/tree/master/Quantum-Dynamics-modules>  
E-CAM Quantum Dynamics Module Documentation ... <http://e-cam.readthedocs.io/en/latest/Quantum-Dynamics-Modules/>  
Read the Docs ... <https://readthedocs.org/>  
Work Package 3 Software Modules ... <http://e-cam.readthedocs.io/en/latest/Quantum-Dynamics-Modules/>  
ReadTheDocs.org ... <http://readthedocs.org>  
QUANTICS ... <http://chemb125.chem.ucl.ac.uk/worthgrp/quantics/doc/index.html>  
ElVibRot ... <https://github.com/lauvergn/ElVibRot-TnumTana>  
here ... [https://www.e-cam2020.eu/wp-content/uploads/2021/03/Workshop\\_scientific\\_report\\_SAW\\_QD\\_2019.pdf](https://www.e-cam2020.eu/wp-content/uploads/2021/03/Workshop_scientific_report_SAW_QD_2019.pdf)

#### Page 3

QUANTICS ... <http://chemb125.chem.ucl.ac.uk/worthgrp/quantics/doc/index.html>  
ElVibRot ... <https://github.com/lauvergn/ElVibRot-TnumTana>  
ElVibRot ... <https://github.com/lauvergn/ElVibRot-TnumTana>

#### Page 4

GitLab service of E-CAM ... <https://gitlab.e-cam2020.eu/>  
Merge-Request of G-CTMQC module. ... [https://gitlab.e-cam2020.eu:10443/e-cam/E-CAM-Library/-/merge\\_requests/267](https://gitlab.e-cam2020.eu:10443/e-cam/E-CAM-Library/-/merge_requests/267)  
readme.rst of G-CTMQC module. ... [https://gitlab.e-cam2020.eu:10443/agostini/E-CAM-Library/-/blob/CT-MQC\\_ecam/Quantum-Dynamics-Modules/modules/G-CTMQC/readme.rst](https://gitlab.e-cam2020.eu:10443/agostini/E-CAM-Library/-/blob/CT-MQC_ecam/Quantum-Dynamics-Modules/modules/G-CTMQC/readme.rst)

#### Page 5

Merge-Request of FBTS-MPI module. ... [https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/-/merge\\_requests/286](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/-/merge_requests/286)  
readme.rst of FBTS-MPI module. ... [https://gitlab.e-cam2020.eu/klparsons/E-CAM-Library/-/blob/FBTS\\_MPI\\_module/Quantum-Dynamics-Modules/modules/FBTS\\_MPI/FBTS\\_MPI\\_readme.rst](https://gitlab.e-cam2020.eu/klparsons/E-CAM-Library/-/blob/FBTS_MPI_module/Quantum-Dynamics-Modules/modules/FBTS_MPI/FBTS_MPI_readme.rst)  
Quantics release 1.3 ... <https://gitlab.com/quantics/quantics>  
Merge-Request of Quantics DD-vMCG MPI/OMP module. ... [https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/-/merge\\_requests/162](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/-/merge_requests/162)  
readme.rst of Quantics DD-vMCG MPI/OMP module. ... [https://gitlab.e-cam2020.eu/thierrytran/E-CAM-Library/-/blob/Quantics\\_DD\\_MPIOMP/Quantum-Dynamics-Modules/modules/Quantics\\_DD\\_MPIOMP/readme.rst](https://gitlab.e-cam2020.eu/thierrytran/E-CAM-Library/-/blob/Quantics_DD_MPIOMP/Quantum-Dynamics-Modules/modules/Quantics_DD_MPIOMP/readme.rst)

#### Page 6

Merge-Request of SHARC-gym module. ... [https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/-/merge\\_requests/160](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/-/merge_requests/160)

readme.rst of SHARC-gym module. ... [https://gitlab.e-cam2020.eu/moritzheindl/E-CAM-Library/-/blob/sharc\\_gym\\_module/Quantum-Dynamics-Modules/modules/sharc\\_gym/readme.rst](https://gitlab.e-cam2020.eu/moritzheindl/E-CAM-Library/-/blob/sharc_gym_module/Quantum-Dynamics-Modules/modules/sharc_gym/readme.rst)

#### Page 7

Merge-Request of ElVibRot-TID-MPI module. ... [https://gitlab.e-cam2020.eu:10443/e-cam/E-CAM-Library/-/merge\\_requests/278](https://gitlab.e-cam2020.eu:10443/e-cam/E-CAM-Library/-/merge_requests/278)

readme.rst of ElVibRot-TID-MPI module. ... [https://gitlab.e-cam2020.eu:10443/achenphysics/E-CAM-Library/-/blob/ElVibRot\\_TID\\_MPI/Quantum-Dynamics-Modules/modules/ElVibRot\\_TID\\_MPI/readme.rst](https://gitlab.e-cam2020.eu:10443/achenphysics/E-CAM-Library/-/blob/ElVibRot_TID_MPI/Quantum-Dynamics-Modules/modules/ElVibRot_TID_MPI/readme.rst)

Merge-Request of ElVibRot-TD-MPI module. ... [https://gitlab.e-cam2020.eu:10443/e-cam/E-CAM-Library/-/merge\\_requests/280](https://gitlab.e-cam2020.eu:10443/e-cam/E-CAM-Library/-/merge_requests/280)

readme.rst of ElVibRot-TD-MPI module. ... [https://gitlab.e-cam2020.eu:10443/achenphysics/E-CAM-Library/-/blob/ElVibRot\\_TD\\_MPI/Quantum-Dynamics-Modules/modules/ElVibRot\\_TD\\_MPI/readme.rst](https://gitlab.e-cam2020.eu:10443/achenphysics/E-CAM-Library/-/blob/ElVibRot_TD_MPI/Quantum-Dynamics-Modules/modules/ElVibRot_TD_MPI/readme.rst)

#### Page 8

E-CAM Pilot Project on Quantum Computing ... <https://www.e-cam2020.eu/pilot-project-ibm/>

here ... <https://e-cam.readthedocs.io/en/latest/Quantum-Dynamics-Modules/index.html#pilot-projects>

PaPIM ... <https://e-cam.readthedocs.io/en/latest/Quantum-Dynamics-Modules/modules/PaPIM/readme.html>

the code's performance analysis ... <https://e-cam.readthedocs.io/en/latest/Quantum-Dynamics-Modules/modules/PaPIM/performance.html>

here ... <https://e-cam.readthedocs.io/en/latest/Quantum-Dynamics-Modules/index.html#papim>

QUANTICS ... <https://www2.chem.ucl.ac.uk/worthgrp/quantics/doc/index.html>

here ... <https://e-cam.readthedocs.io/en/latest/Quantum-Dynamics-Modules/index.html#id7>

ElVibRot ... <https://github.com/lauvergn/ElVibRot-TnumTana>

E-CAM Pilot project on code optimization for exact and linearized quantum dynamics ... <https://www.e-cam2020.eu/pilot-project-on-code-optimization-for-exact-and-linearized-quantum-dynamics/>

here ... [http://pagesperso.lcp.u-psud.fr/lauvergnat/ElVibRot/Tnum-manual-v24.4-09\\_09\\_2013.pdf](http://pagesperso.lcp.u-psud.fr/lauvergnat/ElVibRot/Tnum-manual-v24.4-09_09_2013.pdf)

<https://e-cam.readthedocs.io/en/latest/Quantum-Dynamics-Modules/index.html> ... <https://e-cam.readthedocs.io/en/latest/Quantum-Dynamics-Modules/index.html>

#### Page 9

here ... <https://arxiv.org/abs/2001.03556>

here ... <https://doi.org/10.1039/D0CP01353F>

here ... <https://doi.org/10.1103/PhysRevA.99.052316>

here ... <https://hal.sorbonne-universite.fr/hal-02171352/document>

here ... <https://hal.sorbonne-universite.fr/hal-02298223/document>

here ... <https://www.e-cam2020.eu/tag/quantum-dynamics/>

here ... <https://www.e-cam2020.eu/deliverables/>

ANR ... <https://anr.fr/>

here ... <https://www.e-cam2020.eu/pilot-project-ibm/>

Event website ... <https://www.cecarn.org/workshop-details/330>

Workshop report ... [https://www.e-cam2020.eu/wp-content/uploads/2018/06/Quantum\\_mechanics\\_and\\_electronic\\_structure\\_REPORT.pdf](https://www.e-cam2020.eu/wp-content/uploads/2018/06/Quantum_mechanics_and_electronic_structure_REPORT.pdf)

Event website ... <https://www.cecarn.org/workshop-details/305>

Workshop report ... [https://www.e-cam2020.eu/wp-content/uploads/2018/06/ESDW7\\_Quantum\\_MD\\_REPORT.pdf](https://www.e-cam2020.eu/wp-content/uploads/2018/06/ESDW7_Quantum_MD_REPORT.pdf)

Event website ... <https://www.cecarn.org/workshop-details/179>

Workshop report ... [https://www.e-cam2020.eu/wp-content/uploads/2018/10/ESDW11\\_in\\_quantum\\_dynamics\\_REPORT.pdf](https://www.e-cam2020.eu/wp-content/uploads/2018/10/ESDW11_in_quantum_dynamics_REPORT.pdf)

Event website ... <https://www.cecarn.org/workshop-details/145>

Workshop report ... [https://www.e-cam2020.eu/wp-content/uploads/2021/02/ESDW16\\_Quantum\\_Dynamics\\_REPORT.pdf](https://www.e-cam2020.eu/wp-content/uploads/2021/02/ESDW16_Quantum_Dynamics_REPORT.pdf)

#### Page 11

SAW ... <https://www.cecarn.org/workshop-details/102>

ESDW: Quantum Dynamics ... <https://www.cecarn.org/workshop-details/145>

this location ... <https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/tree/master/Quantum-Dynamics-Modules/modules>

## Citations

- [1] S. Bonella, M. Mališ, B. Courchod, and A. Seitsonen, “Identification/Selection of E-CAM Quantum Dynamics Codes for Development,” Mar. 2018. [Online]. Available: <https://doi.org/10.5281/zenodo.1210416>
- [2] S. Bonella, M. Mališ, A. O’Cais, and L. Liang, “Quantum Dynamics E-CAM modules III,” Mar. 2018. [Online]. Available: <https://doi.org/10.5281/zenodo.1210077>
- [3] C.-Y. Hsieh and R. Kapral, “Nonadiabatic dynamics in open quantum-classical systems: Forward-backward trajectory solution,” *The Journal of Chemical Physics*, vol. 137, no. 22, p. 22A507, 2012. [Online]. Available: <https://doi.org/10.1063/1.4736841>
- [4] —, “Analysis of the forward-backward trajectory solution for the mixed quantum-classical liouville equation,” *The Journal of Chemical Physics*, vol. 138, no. 13, p. 134110, 2013. [Online]. Available: <https://doi.org/10.1063/1.4798221>
- [5] F. Agostini and E. K. U. Gross, *Quantum chemistry and dynamics of excited states: Methods and applications*. Wiley, 2020, ch. Exact factorization of the electron-nuclear wavefunction: Theory and applications.
- [6] S. K. Min, F. Agostini, I. Tavernelli, and E. K. U. Gross, “Ab initio nonadiabatic dynamics with coupled trajectories: A rigorous approach to quantum (de)coherence,” *The Journal of Physical Chemistry Letters*, vol. 8, no. 13, pp. 3048–3055, 2017, pMID: 28618782. [Online]. Available: <https://doi.org/10.1021/acs.jpcclett.7b01249>
- [7] F. Talotta, S. Morisset, N. Rougeau, D. Lauvergnat, and F. Agostini, “Spin-orbit interactions in ultrafast molecular processes,” *Phys. Rev. Lett.*, vol. 124, p. 033001, Jan 2020. [Online]. Available: <https://link.aps.org/doi/10.1103/PhysRevLett.124.033001>
- [8] D. Lauvergnat, “Quantummodellib,” <https://github.com/lauvergnat/QuantumModelLib>.
- [9] J. C. Tully, “Molecular dynamics with electronic transitions,” *The Journal of Chemical Physics*, vol. 93, no. 2, pp. 1061–1071, 1990. [Online]. Available: <https://doi.org/10.1063/1.459170>
- [10] J. C. Tully, “Mixed quantum–classical dynamics,” *Faraday Discuss.*, vol. 110, pp. 407–419, 1998. [Online]. Available: <http://dx.doi.org/10.1039/A801824C>
- [11] G. Granucci and M. Persico, “Critical appraisal of the fewest switches algorithm for surface hopping,” *The Journal of Chemical Physics*, vol. 126, no. 13, p. 134114, 2007. [Online]. Available: <https://doi.org/10.1063/1.2715585>
- [12] E. Marsili, M. Olivucci, D. Lauvergnat, and F. Agostini, “Quantum and quantum-classical studies of the photoisomerization of a retinal chromophore model,” *Journal of Chemical Theory and Computation*, vol. 16, no. 10, pp. 6032–6048, 2020, pMID: 32931266. [Online]. Available: <https://doi.org/10.1021/acs.jctc.0c00679>
- [13] F. Talotta, S. Morisset, N. Rougeau, D. Lauvergnat, and F. Agostini, “Internal conversion and intersystem crossing with the exact factorization,” *Journal of Chemical Theory and Computation*, vol. 16, no. 8, pp. 4833–4848, 2020, pMID: 32633509. [Online]. Available: <https://doi.org/10.1021/acs.jctc.0c00493>
- [14] D. L. C. Pieoroni, E. Marsili and F. Agostini, “Relaxation dynamics through a conical intersection: Quantum and quantum-classical studies,” *To be submitted*.
- [15] R. Kapral and G. Ciccotti, “Mixed quantum-classical dynamics,” *The Journal of Chemical Physics*, vol. 110, no. 18, pp. 8919–8929, 1999. [Online]. Available: <https://doi.org/10.1063/1.478811>
- [16] G. A. Worth, M. A. Robb, and B. L. Lasorne, “Solving the time-dependent schrödinger equation for nuclear motion in one step: Direct dynamics of non-adiabatic systems,” *Mol. Phys.*, vol. 106, pp. 2077–2091, 2008. [Online]. Available: <http://www.tandfonline.com/doi/abs/10.1080/00268970802172503>
- [17] G. A. Worth, “Efficient searching of relevant configuration space: Direct dynamics and the DD-vMCG method,” in *Multi-Dimensional Potential Energy Surfaces*, M. Law, Ed. Daresbury, U.K.: CCP6, 2008, pp. 79–87.
- [18] G. A. Worth, K. Giri, G. W. Richings, M. H. Beck, A. Jäckle, and H.-D. Meyer, “Quantics package, version 1.1,” University of Birmingham, Birmingham, U.K., 2015.
- [19] H.-D. Meyer, U. Manthe, and L. S. Cederbaum, “The multi-configurational time-dependent Hartree approach,” *Chem. Phys. Lett.*, vol. 165, pp. 73–78, 1990.
- [20] S. P. Neville and G. A. Worth, “Conformer-resolved quantum dynamics study of the photodissociation of 3-pyrroline,” *J. Phys. Chem. A*, vol. 118, pp. 11 975–11 986, 2014.
- [21] G. W. Richings and G. A. Worth, “Multi-state non-adiabatic direct-dynamics on propagated diabatic potential energy surfaces,” *Chem. Phys. Lett.*, vol. 683, pp. 606–612, 2017. [Online]. Available: <http://dx.doi.org/10.1016/j.cplett.2017.03.032>

- [22] J. C. Tully and R. K. Preston, "Trajectory surface hopping approach to nonadiabatic molecular collisions: The reaction of  $H^+$  with  $D_2$ ," *The Journal of Chemical Physics*, vol. 55, no. 2, pp. 562–572, 1971. [Online]. Available: <https://doi.org/10.1063/1.1675788>
- [23] M. Beck, A. Jäckle, G. Worth, and H.-D. Meyer, "The multiconfiguration time-dependent hartree (MCTDH) method: a highly efficient algorithm for propagating wavepackets," *Physics Reports*, vol. 324, no. 1, pp. 1–105, 2000. [Online]. Available: <http://www.sciencedirect.com/science/article/pii/S0370157399000472>
- [24] G. Richings, I. Polyak, K. Spinlove, G. Worth, I. Burghardt, and B. Lasorne, "Quantum dynamics simulations using gaussian wavepackets: the vmcg method," *International Reviews in Physical Chemistry*, vol. 34, no. 2, pp. 269–308, 2015. [Online]. Available: <https://doi.org/10.1080/0144235X.2015.1051354>
- [25] S. Gómez, M. Heindl, A. Szabadi, and L. González, "From surface hopping to quantum dynamics and back. finding essential electronic and nuclear degrees of freedom and optimal surface hopping parameters," *The Journal of Physical Chemistry A*, vol. 123, no. 38, pp. 8321–8332, 2019, PMID: 31479265. [Online]. Available: <https://doi.org/10.1021/acs.jpca.9b06103>
- [26] S. Mai, P. Marquetand, and L. González, "Nonadiabatic dynamics: The SHARC approach," *WIREs Computational Molecular Science*, vol. 8, no. 6, p. e1370, 2018. [Online]. Available: <https://onlinelibrary.wiley.com/doi/abs/10.1002/wcms.1370>
- [27] S. Mai, M. Richter, M. Heindl, M. F. S. J. Menger, A. Atkins, M. Ruckebauer, F. Plasser, L. M. Ibele, S. Kropf, M. Oppel, P. Marquetand, and L. González, "Sharc2.1: Surface hopping including arbitrary couplings — program package for non-adiabatic dynamics," [sharc-md.org](http://sharc-md.org), 2019.
- [28] G. A. Worth, K. Giri, G. W. Richings, I. Burghardt, A. M. H. Beck, Jäckle, and H.-D. Meyer, *The QUANTICS Package, Version 1.1*. Birmingham, U.K.: University of Birmingham, 2015.
- [29] D. Lauvergnat, "ElVibRot-TnumTana," <https://github.com/lauvergn/ElVibRot-TnumTana>.
- [30] D. Lauvergnat and A. Nauts, "Torsional energy levels of nitric acid in reduced and full dimensionality with ElVibRot and Tnum," *Phys. Chem. Chem. Phys.*, vol. 12, pp. 8405–8412, 2010. [Online]. Available: <http://dx.doi.org/10.1039/C001944E>
- [31] S. A. Smolyak, "Quadrature and interpolation formulas for tensor products of certain classes of functions," *Dokl. Akad. Nauk SSSR*, vol. 148, no. 5, p. 1042–1045, 1963.
- [32] A. O. Cais, "Esdw technical software guidelines i," Mar. 2016. [Online]. Available: <https://doi.org/10.5281/zenodo.841735>
- [33] L. M. Ibele and B. F. E. Curchod, "A molecular perspective on tully models for nonadiabatic dynamics," *Phys. Chem. Chem. Phys.*, vol. 22, pp. 15 183–15 196, 2020. [Online]. Available: <http://dx.doi.org/10.1039/D0CP01353F>