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Review

Molecular Quantum Dynamics: Recent Perspectives

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Abstract: This review aims to address the current state of the art in the field of Molecular Quantum Dynamics (MQD), where the Schrödinger equation, either in its time-independent or time-dependent form, is solved for the nuclei involved in molecular processes. This is not an exhaustive review of the field, which is far too broad for such an undertaking. Instead, we focus primarily on methods that have enabled the study of larger systems, the size of molecular systems has long been a persistent limitation in the field. We also offer some perspectives, showing that, under certain conditions, it is now becoming possible to treat larger systems(as defined within the scope of this field) in a more systematic way. These advances suggest that MQD's full potential remains to be fully explored.

Key words: molecular quantum dynamics, molecules, quantum, theory.

1. Introduction

This review focuses on the field of Molecular Quantum Dynamics (MQD) [1-5], in which both the electrons and nuclei of a molecular system are treated using a fully quantum-mechanical framework. More specifically, we concentrate on methods in which the Schrödinger equation, in either its time, dependent or time-independent form—is solved for the nuclear degrees of freedom. We do not address semi-classical [6-10] or mixed classical-quantum approaches in this context. With even stronger reason, we do not deal with molecular dynamics (MD) methods, despite their great importance in theoretical chemistry, where the nuclei are treated classically. In particular, despite similarities in terminology, the field of "Molecular Quantum Dynamics" must be clearly distinguished from what is sometimes called "Quantum Molecular Dynamics. In the latter case, the word "quantum" refers to the quantum treatment of the electrons, but, unlike in MQD, the nuclei are treated classically.

The significance of quantum effects in chemistry—such as tunneling, quantum resonances, and quantum interference—is now well established and theoretical chemistry has recently undergone a significant paradigm shift, driven by the more systematic inclusion of quantum nuclear effects in molecular dynamics. Although methods to account for these effects—whether exactly or approximately—were first developed as early as the late 1960s, their critical importance has never been as clearly recognized by the theoretical chemistry community as it is today. This is exemplified by the recent extension of path integral molecular dynamics [11-13] to go beyond the classical treatment of nuclear motion in condensed matter systems [14-16]. For instance, an accurate description of proton transfer mechanisms requires the inclusion of both proton tunneling and the zero-point energy of the molecular system [17]. Similarly, the increasingly acknowledged role of quantum coherence in non-Born–Oppenheimer processes in photochemistry [18,19] has motivated sustained efforts to move beyond traditional approaches such as Ehrenfest dynamics, trajectory surface hopping and more elaborate methods developed by J. Liu and coworkers [20-22], where nuclei are treated (semi)-classically. In practice, however, simulations of nuclear quantum effects remain intrinsically approximate.

Beyond the need for reference methods to assess the validity of more approximate techniques, a fully variational approach that solves the Schrödinger equation is indispensable for a comprehensive description of the emergent quantum phenomena in molecular systems. Molecules are the fundamental building blocks of everyday life. A comprehensive understanding of Molecular Quantum Dynamics (MQD)—and the ability to control it—holds promise for transformative applications, particularly in chemistry, materials science, energy science, biology, and medicine. Such control could enable a fundamental shift from traditional trial-and-error approaches to rational and purposeful molecular design.

Conventional industrial chemical processes are typically governed by macroscopic external parameters, such as temperature, pressure, concentration, solvent choice, or the addition of catalysts. While these methods are effective, they often result in significant energy waste and the generation of large quantities of unwanted by-products. These shortcomings give rise to two major societal challenges: unsustainable energy consumption and environmental pollution. Addressing these issues requires studying chemical processes at their most fundamental level, where quantum effects govern behavior in the microscopic domain. Achieving such an understanding would enable unprecedented levels of control over chemical reactions. In this way, the chemistry of the future—and the innovative applications it enables—will be fundamentally quantum in nature.

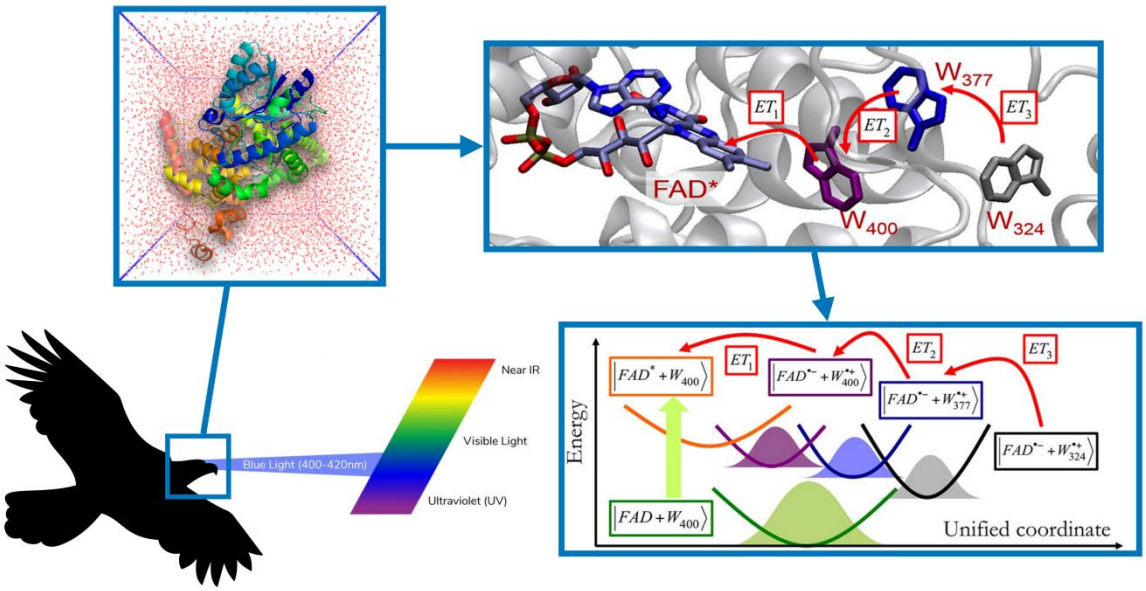

Figure 1. Cryptochromes are a class of flavoproteins involved in regulating circadian rhythms in a wide variety of organisms and are also suspected to play a major role in animal magnetoreception, the ability of migratory birds to detect Earth's magnetic field. Absorption of light induces a radical pair through electron transfers between electronic excited states. Depending on their orientation relative to Earth's magnetic field, these pairs may experience different spin dynamics, leading to distinct physiological responses. Molecular quantum dynamics simulations are now feasible even for such large systems (e.g., cryptochrome in aqueous solvent), enabling a description of quantum coherence, whether vibrational, electronic, or vibronic, in analogous biological processes [50] .

Significant progress has been made on the experimental front, particularly through the development of time-resolved pump-probe laser techniques, which enable the study of chemical processes on the femtosecond timescale (10^{-15} s)—that is, on the timescale of typical molecular vibrations [23-25]. These advances laid the foundation for the emergence of femtochemistry, a field that earned Ahmed Zewail the Nobel Prize in Chemistry in 1999. Femtochemistry allows experimentalists to observe the real-time motion of nuclei, including the breaking and formation of chemical bonds, as well as their geometric rearrangements. More recently, the generation of sub-femtosecond (attosecond) laser pulses has become possible [26-28], opening new avenues for probing electron dynamics on their natural timescale, a possibility that earned Anne L'Huillier, Pierre Agostini and Ferenc Krausz the Nobel Prize in Physics in 2023. These cutting-edge techniques now make it feasible to investigate coupled electronic and nuclear dynamics [29-32]. The ability to control the behavior of both electrons and nuclei has emerged as a cornerstone for understanding a wide range of quantum effects in molecular systems, particularly quantum coherence. Furthermore, through closed-loop laboratory-learning procedures, it is now feasible to design optimal ultrafast laser pulses that guide molecular systems toward a desired target outcome [33-36]. Another major advancement has been the development of methods to orient or align molecules, significantly enhancing the efficiency with which molecular systems can be excited using laser pulses [37-40]. For example, polar molecules in the gas phase can be (partially) aligned or oriented using static electric fields or externally applied fields that vary smoothly in a manner concerted with the molecules' internal motion [41]. The introduction of crossed molecular beam methods, pioneered primarily by Dudley Herschbach and Yuan T. Lee—work for which they were awarded the Nobel Prize in Chemistry in 1986 (alongside John C. Polanyi, recognized for his contributions to chemical kinetics)—has enabled precise measurements of collision cross-sections and reaction rates that are strongly influenced by quantum effects [42,43]. These methods remain powerful even when resonances, such as quantized metastable states, play a dominant role in the scattering process [44-46]. Adding to these capabilities, it is now possible to measure complete vibration-rotation-tunneling (VRT) spectra, even for highly floppy molecular systems such as water clusters [47-49]. Collectively, these experimental advances demonstrate that it is now feasible to detect, quantify with high precision, and even manipulate the majority of quantum effects in molecules.

All of these reasons underscore the essential role of Molecular Quantum Dynamics (MQD). However, despite its considerable promise, the primary limitation of MQD lies in the exponential scaling of the basis-set size with the number of degrees of freedom, which currently restricts practical applications to relatively small systems. Nevertheless, since the author published an initial review on this topic in 2006 [1] and an introductory historical overview of the field in 2014 [2], the progress achieved—particularly in the size of systems that can now be treated—has been remarkable. Even the most recent book dedicated to the Multi-Configuration Time-Dependent Hartree (MCTDH) method, published in 2017 [3], is now somewhat outdated, as significant numerical breakthroughs have been made in the past decade. A new review is therefore timely and necessary. In this work, we do not aim to provide an exhaustive account of all important contributions to the field—such an endeavor would be neither feasible nor desirable. The author respectfully asks colleagues to excuse him should he have inadvertently omitted any of their most significant works. Instead, the main goal of this review is to present the current state of the art in our field to members of both specialized and broader scientific communities. Most importantly, it seeks to demonstrate what MQD can contribute to the future—with the hope of conveying that the

field is still in its infancy, yet it will play a pivotal role in the context of the second quantum revolution, reaffirming that molecules are the fundamental building blocks that shape our world. In particular, the goal of MQD is not only to describe quantum effects in chemistry, but ultimately to exploit those quantum effects— among other applications — in the context of coherent control [51-66].

In section II, we briefly describe the theoretical methodology, highlighting the main numerical bottlenecks. For a much more comprehensive description, we refer the reader to Ref. [3]. This book provides a comprehensive description of the mathematical background as well as an educational presentation of the different aspects of the field, particularly suited for students or young researchers, and includes lab sessions with tutorials illustrating the main topics: infrared and ultraviolet spectroscopy, photodissociation, reactive scattering, and quantum control of molecular processes.

The field of application of MQD is very broad. The goal of this review is not to provide some of the most striking examples of what can be described by MQD, that has already been done in Ref. [2], which includes illustrations of reactive scattering of molecules, photodissociation, surface science, infrared spectroscopy, ultraviolet spectroscopy involving non-adiabatic transitions, photochemistry, attochemistry, and the relation of the field to quantum computing. Instead, we will present the current state of the art in the field in two parts: the applications based on the solution of the time-independent Schrödinger equation in section III or of the time-dependent Schrödinger equation in section.

2. Methodological framework

Solving the full quantum system for a molecular process is well known to be extremely challenging, and we are typically limited to systems involving no more than four atoms. One of the goals of the present review is to demonstrate that it will become possible—in a systematic way—to go far beyond this limitation in the future. In this section, we outline the key technical steps required to model the quantum dynamics of molecular processes: (i)The need for potential energy surfaces (PESs), which must be expressed in a specific form to avoid computationally expensive large mathematical objects; (ii)The selection of an appropriate set of nuclear coordinates and the implementation of the corresponding kinetic energy operator; (iii)The requirement for a suitable primitive basis set, along with the extraction of a much smaller, tractable subspace from this basis to solve the problem efficiently; and (iv)The necessity of employing numerical methods to solve the Schrödinger equation.

2.1. The potential energy surfaces

Solving the Schrödinger equation—whether time-dependent or time-independent—for nuclei in high-dimensional systems requires a vast array of numerical methods. It is impossible to provide an exhaustive list of all the methods developed over the years; instead, we briefly outline the main approaches in this section. As a prerequisite, one must have access to the (possibly coupled) potential energy surfaces (PESs) of the molecular system, typically obtained from quantum chemistry calculations. The quality of the dynamics simulations depends entirely on the quality of these PESs. This dependency is particularly critical in molecular quantum dynamics, where the goal is to accurately describe subtle quantum effects that are observed in cutting-edge experiments. For a long time, the calculation of high-quality PESs was a major bottleneck in the field. However, significant progress has been made in recent years [67]—especially for electronic ground states—thanks, among others, to the systematic application of machine learning techniques [68-94].

Potential energy surfaces for (often coupled) excited electronic states remain significantly more challenging to describe. This challenge is further compounded by the need to diabatize the potentials in order to avoid singular couplings at conical intersections. In many cases, it is necessary (for the calculation of the PESs) to rely on TD-DFT (Time-Dependent Density Functional Theory) calculations. However, accurately describing these potentials—especially the couplings around conical intersections—remains an active area of research in quantum chemistry. In many cases, simplified models such as vibronic coupling models [95-100] or spin-boson models [50,101-103] are employed. These models generally perform well when the system is relatively rigid, even in the emerging field of attochemistry, where many electronic states are involved.

Although some efforts have been made to extend these models to include large-amplitude motions [104-111], there is currently no clear and general framework for formulating models that account for strong deformations of molecular systems involving several coupled electronic excited states. One primary challenge lies in the diabatization of the potentials.

That said, the availability of high-precision global potential energy surfaces (such as those obtained with MRCI (Multireference configuration interaction)) for medium-sized systems in the gas phase is an encouraging development. Here we give just a few examples: Refs. [112-114]. Indeed, it opens up the possibility of making quantitative comparisons between experimental and theoretical results, thereby highlighting quantum effects across a wide range of systems in the near future.

Simply having the potential energy surfaces (PESs) is not sufficient. These PESs often depend on a large number of degrees of freedom, and dealing with the corresponding integrals, matrices, and vectors can quickly become intractable. To address this challenge, a variety of techniques have been developed to avoid working directly with these very large mathematical objects. The most general—and widely applicable—strategy, though not the only one, is to systematically re-express the PESs as a sum of products (SOP) of low-dimensional functions [67]. This approach allows, for example, the calculation of a multidimensional matrix to be reformulated as a sum of products of low-dimensional matrices, which are much more manageable [115]. Even though numerical strategies exist to treat rather large systems without resorting to a SOP form for the Hamiltonian, avoiding this form becomes very difficult for large systems [116].

Of course, such a re-expression requires significant effort, and developing a general algorithm that can be applied to large systems remains a considerable challenge [67,117-129]. As we will see below, however, significant progress has recently been made in this area—particularly in the context of the MCTDH method [130].

2.2. The choice of the coordinates and the kinetic energy operator

A second critical step in molecular quantum dynamics is the selection of an appropriate coordinate system to describe nuclear

motion—a choice that is far from trivial. While rectilinear coordinates (e.g., Cartesian coordinates or the normal modes of traditional spectroscopy [131]) may suffice in certain cases, many molecular processes—particularly those involving large-amplitude motions such as rotation, torsion, isomerization, or collisions—require curvilinear coordinates. These are coordinates that cannot be expressed as linear combinations of Cartesian coordinates (typically involving angles) and may be essential for accurately describing the system's dynamics. The challenge is not that rectilinear coordinates fail to describe the process; rather, they introduce extreme numerical coupling between degrees of freedom, often rendering the numerical solution of the Schrödinger equation intractable. In such cases, the choice of a coordinate system tailored to the physical process is not merely beneficial—it is fundamental to enabling a meaningful solution. Naturally, curvilinear coordinates result in significantly more complex kinetic energy operators (KEOs) for the nuclei, which must be carefully derived and implemented in the computational codes used to solve the Schrödinger equation for the nuclear motion.

Ideally, one might envision identifying the "optimal coordinates" for a given molecular process. Here, we define "optimal coordinates" as those that enable the solution of the Schrödinger equation to converge using the smallest possible number of basis set functions. Some studies have demonstrated that convergence can be significantly improved by "optimizing" the coordinate system [132,133]. In practice, however, the situation is more nuanced. Even for a very specific process, the "optimal" coordinates depend on the energy regime explored by the wavefunction. For instance, consider a scenario where a wavepacket splits into multiple components across different electronic states. In such cases, the optimal coordinates may differ not only between electronic states but also across the various energy domains spanned by each component. Moreover, coordinates that yield optimal convergence for the wavefunction may lead to poor convergence when expressing the potential in a sum-of-products (SOP) form. They can also result in kinetic energy operators (KEOs) with an excessive number of terms, or even introduce so-called singularities [3]—terms that tend to infinity for certain coordinate values (e.g., the spherical coordinates φ and θ when θ approaches 0 or π). Such singularities may necessitate the use of multidimensional basis sets, such as spherical harmonics [134], which can further complicate the numerical solution of the Schrödinger equation. In practice, one must strike a balance among these competing constraints. The goal is to select coordinates that are well-suited to the physical problem at hand and that allow the calculation to converge efficiently—without attempting to optimize the coordinate system at all costs.

For processes involving large-amplitude motions, one family of curvilinear coordinates that has proven to offer a good compromise for many applications is the family of so-called polyspherical coordinates [135-140]. These are a generalization of spherical coordinates, offering significant flexibility in their definition. They include, for example, Jacobi coordinates—commonly used for describing collisions or isomerization processes—as well as valence coordinates, which directly represent interatomic distances. A key advantage of polyspherical coordinates is their ability to define localized subsystems, allowing coordinates to be tailored to specific regions of the molecule. This helps avoid undesired couplings between atoms that are physically distant, a problem often encountered with standard spherical coordinates. Another major benefit is that polyspherical coordinates always yield a kinetic energy operator (KEO) in a SOP form, expressed as a combination of one-dimensional operators. This property simplifies both the theoretical formulation and the numerical implementation. Other coordinate systems are also widely used. For example, hyperspherical coordinates [141] are popular for collision dynamics, particularly in the context of ultracold chemistry [142]. Additionally, various other curvilinear coordinates have been employed to address specific challenges. Let us just give two examples: coordinates that preserve symmetry in systems such as $H + CH_4$ collisions [143]; coordinates designed to minimize coupling between rotation and internal vibration for each water molecule in the water dimer [144].

Deriving the kinetic energy operator (KEO) was particularly challenging in the early stages of molecular quantum dynamics [145-151]. For large systems, one solution has been to compute the functions involved in the KEO numerically for given geometries [152-155]. The advantage of this approach is that the KEO can be obtained for any set of coordinates. However, it does not yield the KEO in a sum-of-products (SOP) form.

A major improvement came with the development of software—such as the TANA program [156-158]—capable of providing an analytical KEO for polyspherical coordinates. As previously mentioned, polyspherical coordinates offer several advantages. The automated procedure implemented in such software allows for the treatment of even very large systems involving large-amplitude motions, as we will discuss further below.

2.3 Solving the Schrödinger Equation for the nuclei

In addition, the wavefunction must be expanded in a mathematical basis set—often called a primitive basis set. In many cases, though not exclusively, a dual finite-basis-representation (FBR)/discrete-variable-representation (DVR) [159-165] scheme is employed. The FBR is more adapted to the derivative operators that appear in the KEO, the DVR to the multiplicative operators such as the PES. These approaches treat the KEO efficiently within FBR or the PES efficiently within DVR. However, the number of primitive functions grows exponentially with the number of degrees of freedom. Since the early days of molecular quantum dynamics (MQD), a key advance has been to extract from the primitive basis a much smaller active subspace in which the Schrödinger equation can be solved [166-168]. This subspace is often—but not always—formed from contracted functions, which are linear combinations of the primitive functions. In the following sections, we will give a few examples of algorithms to extract such an active subspace.

Finally, one must solve the Schrödinger equation. For the time-dependent case, numerous numerical integrators have been implemented [169-172]. Recently, the first attempts have been made to accelerate wavepacket propagation using machine learning [173]. For the time-independent case, the Hamiltonian matrix is diagonalized [67]. Because the resulting matrix can be very large—even in a contracted basis—iterative methods like Lanczos [174-180] or Davidson [181-185] are typically preferred.

3. Applications using a time-independent method

3.1 Rovibrational spectroscopy

The independent approach is typically reserved for high-accuracy calculations, such as highly resolved spectroscopy or low-

temperature collisions where quantum resonances must be described precisely. Within MQD, we employ ab initio methods in which all calculations are performed starting from ab initio potential energy surfaces (PESs). The computation of infrared (IR) spectra has been the subject of extensive study because IR spectroscopy is central to molecular characterization, including in astrophysical contexts for elucidating atmospheric chemistry [186,187]. In addition, these high-accuracy calculations serve as essential benchmarks for other methods in MQD, enabling verification of the correctness of implementations [188]. As noted, one challenge lies in the size of the basis set required to solve the time-independent Schrödinger equation and obtain the rovibrational eigenstates and eigenvalues that correspond to the transitions observed in spectra. An active subspace must first be extracted to drastically reduce the problem size. A particularly successful tool in this domain is the MULTIMODE code developed by J. Bowman and coworkers [189-192] Inspired by methods that have been proven to be very successful in quantum chemistry, the code exploits Vibrational Self-Consistent Field (VSCF) or Vibrational Configuration Interaction (VCI) algorithms that optimize an intermediate basis set. Here, a variational principle for the time-independent Schrödinger equation is applied to obtain intermediate functions that converge to the eigenstates with a much smaller number of functions than required by the primitive basis set. MULTIMODE has been used to systems as large as C_2H_4, CH_3OH, CH_3NO_2, CH_3CHOO. This approach has also yielded important insights into larger, highly flexible systems such as water clusters or protonated water clusters, albeit in reduced dimensionality and often combined with Monte Carlo simulations [193-198]. VSCF and VCI methods have also been applied to the study of IR spectroscopy by the group of Pau, in the MOLPRO package [199-204], by Liévin and coworkers [205], etc.

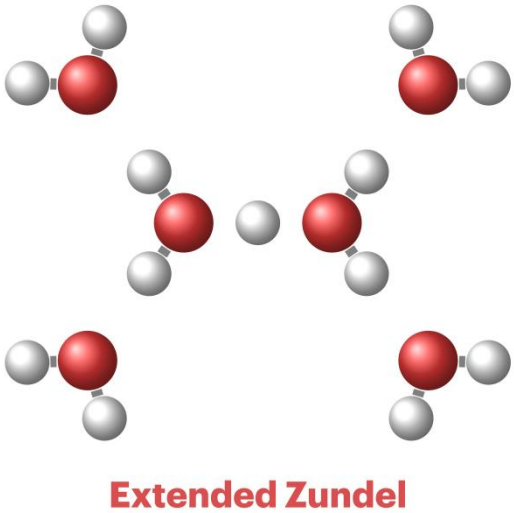

Figure 2. In Ref. [227], the infrared spectrum of the extended Zundelcation was calculated using a full quantum approach in 51 dimensions, showing excellent agreement with experimental data. The system exhibits strong anharmonicity and significant vibrational coupling. It serves as a key model for understanding the behavior of protons in aqueous environments.

When the molecular systems are very floppy, meaning they exhibit many large-amplitude motions—as in van der Waals dimers, water clusters, or even more rigid but very high-energy systems—the computation of rovibrational eigenstates remains extremely challenging, due to a very high anharmonicity and very strong couplings between the modes of vibration, and is still typically limited to systems of at most six atoms. Of course, as in quantum chemistry, combined variational and perturbative methods have also been systematically adopted—for example, by E. Sibert and coworkers [206-209]—allowing, under specific conditions, the treatment of larger systems than usual (more than 15 atoms). A strategy based on a coupled cluster method, again inspired from quantum chemistry, has been developed by O. Christian and coworkers to calculate vibrational eigenstates in a systematic way [210-213] including a description in curvilinear coordinates [214-216]. Other codes have been developed to compute infrared spectra in a systematic fashion, often limited to small molecules, but capable of accessing very high energies and extracting information from experimental data that can be used with the GENIUSH and MARVEL codes developed by A. G. Császár and coworkers [217-226].

Many algorithms have been developed to treat ever larger systems and to tackle the description of systems involving motions of large amplitude. In this context, the group of T. Carrington Jr. has played a leading role in devising a wide range of numerical strategies [180,228-250] It does not make sense to try to list all these strategies. Let us simply mention the approach based on a separate contraction of the angular and radial basis sets (by fixing the other degrees of freedom, radial or angular), combined with the Lanczos algorithm. This approach has been used successfully to converge the first eigenstates of the very fluctional system CH_5^+ [251] and, more recently, the water dimer [144,252], even in the region of the OH stretching modes, which was still the subject of considerable controversy. Among the different numerical methods tested by T. Carrington et al., the Smolyak scheme [253] has been exploited by other groups such as D. Lauvergnat or Y. Scribano et al. to calculate vibrational eigenstates in systems such as H_2 embedded in very large systems such as an sII clathrate hydrate. Similar strategies for contracting the primitive basis—while fully exploiting molecular symmetry and optimizing the discrete variable representation (DVR) with respect to the potential energy [Potential-Optimized DVR (PODVR) [254]]—have recently been developed by P. Felker, Z. Bačić, and coworkers. These advances have enabled, for the first time, the calculation of all the fundamental eigenstates of dimers such as the HCl–H_2 O dimer [255-264], as well as several trimers including the intermolecular bending states and tunneling splittings of the water trimer [265-271]. All of these works became feasible thanks to the systematic exploitation of the permutation–inversion group symmetry of the molecules [272]. This symmetry-based approach enables calculations to be performed separately within each irreducible representation, thereby reducing computational cost because the Hamiltonian matrix is smaller in each representation. The same concept—contracting the basis set and exploiting symmetry—was later employed by M. Rey and coworkers to compute rovibrational eigenstates at very high energies in systems such as methane (CH_4), trifluoromethane (CHF_3), and other molecules of major greenhouse-gas or astrophysical importance [273-282]. A systematic use of symmetry has also allowed van der Avoird and coworkers to calculate the tunneling splittings in very difficult problems such as the benzene or the ammonia dimers [283,284].

To provide a concrete example of what can be achieved—and of the challenges that the field still faces—let us consider one case. There is still no precise understanding of the experimental infrared

spectra in the 2000-4000 cm^{-1} region for water clusters (such as the dimer, trimer, tetramer, etc.): see Refs. [252,285] for the ongoing difficulties in assigning the spectrum of the water dimer. Achieving such an understanding is essential for correctly describing the hydrogen-bond network in liquid water and the crystallization process of ice.

3.2 Molecular collisions

To conclude this section, we mention the time-independent formulation used to calculate cross sections for molecular collisions [286].

Unlike time-dependent methods, this approach is well suited for treating collisions that exhibit very dense quantum resonances with long lifetimes, as well as collisions at ultracold temperatures [142,287-293]. However, this method remains limited in dimensionality: it is typically applicable to systems with only a few atoms (often three) for reactive collisions, and to somewhat larger systems for inelastic collisions — for example, using the MOLSCAT program [294]: see Ref. [295] and [296] for a comparison between time dependent and time-independent methods for inelastic collisions.

4. Applications with time-dependent methods

4.1 General strategies in quantum dynamics

The time-dependent approach is fundamentally different from—but highly complementary to—the time-independent one. Of course, due to the Heisenberg uncertainty principle, a time-dependent approach is not well suited for achieving very high accuracy in energy. It is possible to simulate the evolution of a wave packet by expanding it in the eigenstate basis of the system and applying the time-evolution operator. This approach has been applied to the study of intramolecular vibrational energy redistribution (IVR) in small molecules, in conjunction with their infrared spectroscopy, by, for instance, R. Marquardt, M. Quack and coworkers [297-306]. However, such an approach requires two conditions: not only must the eigenstates be converged to sufficient accuracy, but also all those that play a role in the dynamics must be included in the simulation. Even for the study of IVR, the number of eigenstates can easily become extremely large even for small molecules [307].

By contrast, time-dependent methods that numerically solve the Schrödinger equation have a key advantage: they can describe strong quantum mechanical processes—involving very large numbers of eigenstates or quantum resonances—without requiring knowledge of each eigenstate individually.

Time-dependent methods have been used very successfully in many areas of molecular quantum dynamics (MQD), particularly for non-elastic and reactive scattering. These methods often make use of the aforementioned PODVR basis set to reduce the number of functions required. Among others, the seminal works of the research groups led by H. Guo [308-331] and D.-H. Zhang [45,46,332-341] can be highlighted in many fields of applications in particular in the context of reactive scattering. In the latter context, one could also highlight the works of the groups of G.-J. Kroes (Refs. [342-351]) or B. Jackson ([352-359]). As for the time-independent approaches, these time-dependent methods are typically limited to systems of six atoms in full dimensionality [360] (see also the very recent simulation for a system with seven atoms an excellent agreement with experiment but with reduced dimensionality [361]).

To clearly illustrate the current limits, consider the accurate description of C–H bond breaking of CH_4 on a metal surface such as Ni(111)—a fundamental process for industrial applications. This problem is not yet solved. It is particularly challenging because all internal vibrational modes must be described; in experiments, lasers excite these modes to probe the role of each one [362-364]. Moreover, C–H bond rupture involves strong correlations among many motions, notably the orientation and thus the rotation of the molecule. On top of that, non-adiabatic effects may play a role, and the motion of the substrate atoms must be included in the simulation.

Time-dependent quantum simulations find application across a remarkably broad spectrum, including quantum control, photochemistry, and infrared/UV spectroscopies, to name just a few. It would be impractical to attempt an exhaustive enumeration of the works that employ such time-dependent approaches; instead, we will focus on the central question: is it realistic to expect that we can tackle much larger systems in the future? This question is all the more timely because, as mentioned above, high-quality potential energy surfaces (PESs) are becoming increasingly available, in part because of the use of machine learning.

4.2 The Multi-Configuration Time-Dependent Hartree (MCTDH) approach

The most widely used method that has proven capable of treating large systems across various fields of molecular quantum dynamics (MQD) is the Multi-Configuration Time-Dependent Hartree (MCTDH) method, originally developed by H.-D. Meyer, U. Manthe, and L. Cederbaum [365,366] (see also references [367] and [368] for pioneering works). As for the MULTIMODE code, a variational principle is used to derive a contracted basis set—significantly smaller than the primitive basis set. The key difference lies in the fact that this variational principle is applied to the time-dependent Schrödinger equation. In other words, a set of functions called single-particle functions (SPFs) are time-dependent and expressed in terms of the primitive basis functions. These SPFs are optimized using the Dirac–Frenkel variational principle [115]. The time dependence of the SPFs is crucial, as it allows them to adapt dynamically during the propagation of the wave packet, in response to the evolving physical situation. MCTDH can be viewed as a time-dependent analogue of CASSCF or MCSCF method, but applied to the nuclear degrees of freedom. In this framework, the active subspace evolves over time, reflecting the changing physical conditions experienced by the molecular system.

For MCTDH, several tools and frameworks have been developed. H.-D. Meyer and coworkers have implemented an extensive toolbox of numerical techniques in the Heidelberg MCTDH package [369]. This includes not only the MCTDH algorithm itself, but also a wide range of other numerical methods originating from the molecular quantum dynamics (MQD) community — such as libraries for primitive basis sets, integrators, various operators, and numerous analysis programs, among others [115,370]. The approach developed by H.-D. Meyer and coworkers makes systematic use of the sum-of-products form of the Hamiltonian. To support this, the package includes several programs that can transform potential energy surfaces into this form, if necessary [67,117-129]. In this approach, the single-

particle functions (SPFs) can depend on several degrees of freedom (typically 1 to 4 degrees of freedom, in order to keep the primitive basis set of the corresponding "combined mode" from becoming too large). The package has been applied to a very broad range of systems, typically involving up to 10 atoms in full dimensionality. One particularly successful application was the calculation of ultraviolet (UV) spectra involving conical intersections — that is, cases of very strong non-adiabatic coupling between electronically excited states. An extensive use has been made of vibronic coupling models, such as those developed by H. Köppel and coworkers [371-373]. One advantage of these models is that, when expressed in normal coordinates, they automatically adopt the sum-of-products form. As a result, experimental spectra — such as those of pyrazine (a 10-atom molecule) — have been accurately reproduced using full-dimensional wave packet propagations (24 degrees of freedom for pyrazine [96,374], see also Refs. [98] for additional examples). The Heidelberg package was further included in the more general package QUANTICS by G. A. Worth and coworkers (see below) [375].

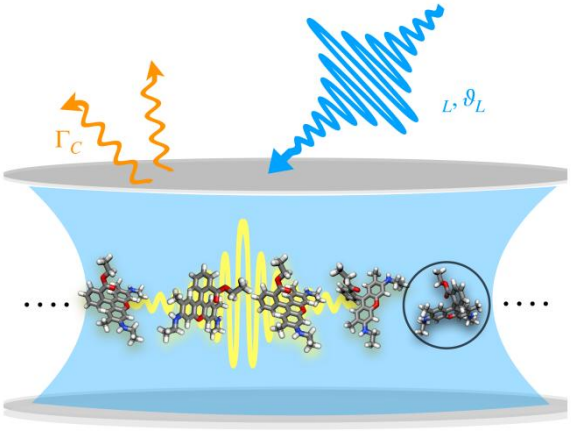

Figure 3. In Ref. [382], the study of exciton-polaritons in microcavities has shown that they can lead to an efficient energy transport in organic materials. The optical Fabry-Pérot microcavity is filled with Rhodamine chromophores, in which all molecular transitions dipole moments are aligned with the cavity polarization direction. The quantum simulations involved around 500 degrees of freedom.

Another approach, developed by U. Manthe and coworkers, is based on a time-dependent optimized DVR or Correlated DVR (CDVR) [376]. This method offers the major advantage of avoiding the need for a sum-of-products (SOP) form of the Hamiltonian operator. However, it requires evaluating the potential at each time step during propagation and does not permit a natural implementation of mode combination. Among many applications, U. Manthe was able, for the first time, to describe reactive scattering with 6 atoms in full dimensionality, much earlier than was possible with more standard approaches [377-381].

Several variants of MCTDH have since been developed, significantly broadening the range of applications. One of the most important advances was the introduction of the Multi-Layer Multi-Configuration Time-Dependent Hartree (ML-MCTDH) approach by H. Wang and M. Thoss [383-387]. In this approach, multidimensional time-dependent single-particle functions (SPFs) are themselves expressed in terms of lower-dimensional SPFs, which are also optimized using the variational principle — rather than being expressed directly in the primitive basis set. This hierarchical structure allows for the inclusion of multiple layers, resulting in a highly flexible representation and a very compact form of the wave packets. The original formulation of ML-MCTDH has been further refined by U. Manthe [388] and implemented by O. Vendrell and H.-D. Meyer in the Heidelberg package [389]. It has been demonstrated, as a proof of principle using model systems, that ML-MCTDH can handle systems with up to thousands of nuclear degrees of freedom [390]. For such large systems, however, a proper regularization of the equations of motion in ML-MCTDH must be implemented [390-392]. In any case, the advent of the ML-MCTDH approach has marked a major advancement. ML-MCTDH is currently been used in many groups (along with more standard approaches for small systems). Let us just mention a few examples: the S. Mahapatra and coworkers to study nonadiabatic quantum molecular dynamics for reactive scattering or dynamics of molecules on coupled electronic states after photoabsorption [393-413], A. Viel and coworkers in the same field [81,83,414,415], A. Kuleff and coworkers in the novel field of attochemistry where many coupled electronic states are involved [416-423] and S. Guérin, M. Sala and coworkers [424-427] or N. Henrinksen and coworkers [428-433] for quantum control of chemical processes. The program developed in Bielefeld has also enabled U. Manthe and coworkers to play a key role in the quantum description of chemical reactions, particularly for reactions involving more than four atoms [377-381,434-449].

However several technical challenges remain for the use of ML-MCTDH for very large systems. These include the availability of the potential energy surfaces (PESs), their refitting if necessary, the choice of the tree structure in ML-MCTDH, the selection of coordinates, and the implementation of the corresponding kinetic energy operators (KEOs), among others. Before discussing how these issues can be addressed within the standard ML-MCTDH framework, let us first present the other variants of MCTDH. One line of development has been to constrain the single-particle functions (SPFs) to retain a Gaussian shape throughout the propagation. This yields the Gaussian MCTDH (G-MCTDH) variant introduced by I. Burghardt ad coworkers [450-453](see also Refs. [454] and [455] for recent applications of G-MCTDH for large systems). A key advantage is that it enables the definition of trajectories associated with the Gaussian functions, thereby facilitating on-the-fly potential energy evaluation in a manner analogous to popular surface-hopping methods in photochemistry [19]. However, G-MCTDH not only incorporates nuclear quantum effects but, unlike other Gaussian-propagation approaches [19], is also variational: if a sufficient number of Gaussian functions are propagated, the method converges to the correct result. The G-MCTDH approach has since been generalized to a multi-layer formulation, analogous to standard MCTDH [456-461]. This framework has enabled G. A. Worth and coworkers to develop a comprehensive set of techniques based on G-MCTDH for studying the photochemistry of large systems, including those involving strong non-adiabatic processes [462-473]. In particular, G. A. Worth and coworkers compute the potential energy on the fly using the DD-vMCG method — the Direct Dynamics Variational Multi-Configurational Gaussian approach. In practice, this allows for local diabatization to be achieved. The method is implemented in the Quantics software package [375,474]. Additionally, the package enables the collection of a large number of potential energy values at different geometries, which can then be used to fit

potential energy surfaces (PESs).

However, it should be emphasized that the philosophy behind these on-the-fly approaches begins to diverge—albeit in a complementary way—from all previously described grid-based methods (so called because they employ a primitive basis set, often DVR). In particular, while the DD-vMCG method is inherently variational, the number of Gaussian functions is often limited by the cost of computing the potential on the fly. Moreover, the employed quantum chemistry methods generally cannot be very demanding (such as MRCI). In other words, this approach emphasizes obtaining a qualitative picture of the quantum dynamics, especially in photochemistry. This limitation aligns with the current difficulty of achieving highly accurate quantum chemistry descriptions for coupled electronic states in large molecules. As noted earlier, the two paradigms are complementary: grid-based approaches—such as those implemented in the Heidelberg MCTDH package—are warranted only when the PESs are of very high quality and when accurate experimental data are available that demand interpretation at that level of precision. Within the current state of the art, achieving such high-quality potential energy surfaces (PESs) is possible for large systems only in the electronic ground state.

In the further development of G-MCTDH, significant effort has been devoted to treating even larger systems, in particular those coupled to a very large bath that acts as a reservoir for energy transfer. The multiconfiguration time-dependent Hartree (MCTDH) method has naturally been generalized to the propagation of density operators [475-477], since the latter allow, among other things, for the description of open quantum dynamical systems using the formalism of Lindblad operators from density matrix theory. However, the propagation of density operators is more computationally expensive, and alternative strategies have been adopted. D. Picconi, I. Burghardt, and coworkers have developed several approaches to incorporate the effects of dissipation within MCTDH. For instance, they have combined MCTDH (specifically a hybrid of MCTDH and G-MCTDH) with a treatment of dissipation and decoherence based on the nonstochastic open-system Schrödinger equations [478,479]. This approach was partly inspired by earlier work by L. Joubert Doriol and A. Izmaylov and coworkers [480,481]. A more recent approach, implemented in the Heidelberg MCTDH package by J.-C. Tremblay and coworkers [482-484], describes multidimensional stochastic dissipative quantum dynamics using MCTDH with a Lindblad operator. It employs the Monte Carlo wave packet (MCWP) algorithm, which was shown to be equivalent to the reduced-density-matrix propagation in the Lindblad form under specific conditions [485-487]. Such an approach can be essential, in particular, for describing the motion of molecules on surfaces (see for instance Ref. [488-490]).

As an illustrative example of the efficiency to transfer the numerical methods in MQD, especially MCTDH, outside their initial field of research we mention the fact that MCTDH has been generalized to describe systems made of indistinguishable particles. This was done in particular to describe the physics of Bose-Einstein condensates or strong-field electron dynamics. Thus, MCTDH-F [491-493] for fermions and MCTDH-B [494-498] for bosons or, together, MCTDH-X [499-503] approaches were developed. To preserve the exchange symmetry of indistinguishable particles and to leverage the advantages of the Multi-Layer MCTDH framework, a second-quantized formulation of the latter has been proposed [387,504]. We refer the reader to the major review by O. Alon and coworkers for a comprehensive description of these methods [505].

MCTDH has been adapted by H.-D. Meyer and coworkers in the Heidelberg package to solve the time-independent problem [122,184,185,506]. The method can then be viewed as a time-independent MCSCF approach for the nuclei. This approach is particularly useful, as it can be used as a black-box tool within the package even for floppy systems. However, the numerical efficiency of this method does not match that of the most recent approaches described in the previous section, for example, the methods developed by T. Carrington and coworkers for the water dimer. This time-independent version of MCTDH has also been exploited by U. Manthe and coworkers, for isntance for the calculation of the tunneling splittings of vibrational states in malonaldehyde [507-509].

4.3 State of the art depending on the form of the Hamiltonian operator

Let us now return to the "standard" ML-MCTDH approach and describe precisely the current state of the art in terms of what is achievable. Two distinct cases must be considered:

- First, the Hamiltonian appears in sum-of-product form with respect to the coordinates used for the dynamics — that is, the Hamiltonian is, in most cases, derived from a model describing a specific process.
- Second, the Hamiltonian includes one or several ab initio potential energy surfaces (PESs), which may also involve couplings between those surfaces, as well as dipole moment or polarizability surfaces.

In the second case, when using the Heidelberg program, these PESs must be refitted into sum-of-product form.

Independently of the case, a major breakthrough has been achieved very recently by D. Mendive Tapia and coworkers [510] by devising quantitative criteria for choosing the tree structure in ML-MCTDH (see also the very recent review summarizing the choice of the tree structure for reaction dynamics by Q. Meng and coworkers [511]). Indeed, when many degrees of freedom are involved, a large number of possible tree topologies may exist, and the convergence of the propagations strongly depends on the choice of the tree — just as it also depends significantly on the choice of the coordinate system. In practice, one evaluates the coupling between the coordinates for a given wave function and a given Hamiltonian operator, and the coordinates are grouped accordingly. Obviously, strongly coupled coordinates should appear close to one another in the tree, rather than being placed in distant branches.

Now, in the first case, ML-MCTDH is being used for systems with hundreds of nuclear degrees of freedom, making it possible to converge the quantum dynamics — even for biological processes in solution [50,512,513]. This opens the door to obtaining a rigorous description of quantum effects, such as quantum coherence, in biological systems, which remain the subject of ongoing debate [514.515]. However, it must be emphasized that, even when the nuclear quantum dynamics is fully converged (which can be done with ML-MCTDH and by increasing the number of degrees of freedom until the dynamics does not change any more [50]), the quality of the results depends strongly on the quality of the underlying model and, equally importantly, on the accuracy of the quantum chemistry calculations. For large organic systems, and even more so for biological systems involving coupled electronic

excitations, the accurate description of the electronic potentials remains challenging. As previously mentioned, on-the-fly methods such as DD-vMCG may be preferred when a high-quality quantum chemistry description cannot be guaranteed.

However, the model can be of very high quality in describing the underlying quantum process, and ML-MCTDH therefore provides the ideal tool for converging the dynamics. As a pertinent illustration, we can mention the very recent quantum dynamics simulation of exciton–polariton transport — involving around 500 degrees of freedom — carried out by O. Vendrell and coworkers [382]. Other examples include the studies of charge-transfer excitons by I. Burghardt and coworkers in several systems, encompassing up to 25 electronic states and 875 vibrational modes [516-521].

At this level, it is worth highlighting a systematic comparison published in Ref. [522] between ML-MCTDH and the multi-electronic-state path integral molecular dynamics (MES-PIMD) approach developed by J. Liu and coworkers. The two methods show excellent agreement for Spin-Boson models with a very large number of degrees of freedom. ML-MCTDH appears more efficient at zero or low temperatures, while MES-PIMD offers greater computational advantages at relatively high temperatures. ML-MCTDH enables the study of processes in the condensed phase, but it is anticipated that path integral-based methods, combined with suitable approximations, will become better suited for systematic investigations of the condensed phase—particularly the liquid phase—in the future. While this review does not focus on path integral-based methods, we emphasize again the complementarity between the molecular quantum dynamics methods discussed here and those based on path integrals.

In the second case, i.e. when using ab initio PESs, another major breakthrough has been the development and implementation in the Heidelberg package of the canonical polyadic decomposition using Monte Carlo (MCCPD) method by M. Schröder [130] , which enables the refitting of potential energy surfaces (PESs) into sum-of-products form. Indeed, this is currently the only method capable of systematically refitting large PESs. As an illustration, we mention the calculations of the infrared spectra of the most important protonated water clusters, performed by O. Vendrell and coworkers: the Eigen cation in 33 dimensions (13 atoms) [523] and, more recently, the extended Zundel cation in 51 dimensions (19 atoms) [227]. These systems are highly anharmonic and exhibit very strong coupling between many vibrational motions. The simulations made use of very high-quality PESs of the electronic ground state, along with dipole moment surfaces, all in full dimensionality. A simulation in normal coordinates was not feasible — for such systems, normal coordinates do not even allow convergence of the vibrational ground state. Instead, well-chosen polyspherical coordinates [140] were employed to describe these systems, which exhibit many large-amplitude motions. The kinetic energy operators (KEOs), which include thousands of terms, were automatically generated in an input file read by the Heidelberg package, using the TANA program [156-158]. As previously mentioned, this automatic implementation represents a major advantage when dealing with very flexible (floppy) systems. The simulations yielded spectra in excellent agreement with experimental data, along with a comprehensive description of the various spectral peaks and of the couplings within the system — in particular, the coupling between proton motion and the different water molecules embedded in a hydrogen-bonded network. The agreement between experiment and theory also validates the quality of the PESs, as well as the implementation of all the technical methods used in the calculations — including the MCCPD refitting procedure.

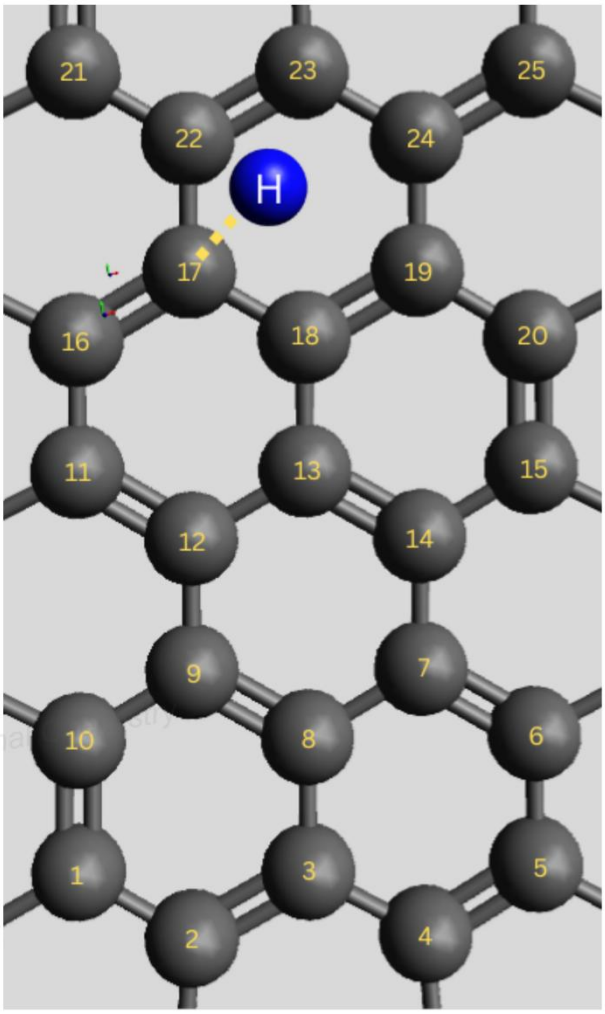

Figure 4. In Refs. [524] and [525], 75 dimensional wavepacket simulations were carried out to describe hydrogen scattering on graphene with different ingoing kinetic energies and angles. A cell of 24 Carbon atoms was included and treated periodically. The simulations employed an *ab initio* potential energy surface (PES) based on Neural Network DFT (NN-DFT) calculations. The goal was to interpret experimental results such as those in Ref. [526]. The description of the system is particularly challenging since the H atom can reach all the parts of the surface and can create C-H bonds leading to a strong anharmonicity of the Carbon motions. The system is a prototype to analyze the formation of a C-H bond at the elementary level and to understand the transfer of energy from a particle with a surface.

Recently, a similar strategy — except that Cartesian and normal coordinates were used instead of polyspherical coordinates — was employed to study quantum molecular dynamics for H-atom scattering from graphene, involving 75 dimensions (25 atoms) by L. Shi and coworkers [524.525,527]. This represents the largest quantum simulation with an ab initio potential energy surface (PES) with ML-MCTDH to date. The excellent agreement between the quasi-classical simulations and the quantum simulations in the high-energy regime further confirms that the full machinery of

techniques — including the refitting with MCCPD — is well controlled. There is no reason to believe that this global strategy could not be applied to many other systems, including even larger ones. However, it must be emphasized that MCCPD is not fully variational, and therefore cannot guarantee very high accuracy (for example, on the order of 1 cm^{-1}). In addition, ML-MCTDH, like any quantum time-dependent method, becomes computationally very expensive for long propagations. For the H/graphene system, the propagations covered a few hundred femtoseconds, and for the protonated water clusters, they extended to a few picoseconds. ML-MCTDH is well suited for strongly quantum but fast processes. This limitation applies to all time-dependent methods, but it is even more pronounced for ML-MCTDH, since if one aims to describe very fine details, the advantage of the method's compactness is quickly lost.

4.4 Link to Density Matrix Renormalization Group (DMRG)

To conclude, let us compare ML-MCTDH with the widely used Density Matrix Renormalization Group (DMRG) method. For an in-depth discussion of the comparison between these two approaches, we refer the reader to a recent and excellent publication by H. Larsson [528] and the references therein. We have not discussed DMRG until now because it originates from the field of condensed matter physics and has not yet been applied to molecular quantum dynamics (MQD). Originally, DMRG was developed to calculate eigenstates, particularly the ground state of one-dimensional chains. For converging the ground state of a very long one-dimensional chain, DMRG is naturally more efficient than ML-MCTDH [529] DMRG was specifically optimized for this case, whereas ML-MCTDH is fundamentally a time-dependent method. Subsequently, DMRG has been extended to handle multi-dimensional eigenstates and to perform time-dependent simulations, giving rise to Time-Dependent DMRG (TD-DMRG). As explained by H. Larsson, the fact that the two methods were developed by two different scientific communities — using different conventions — has led to very different mathematical "languages", which can obscure their underlying similarities. While the ways of optimizing the functions may differ, the overall structure of ML-MCTDH and TD-DMRG is identical. As shown by H. Larsson, the two methods share a common mathematical foundation. In particular, the Matrix Product State (MPS) corresponds to a very specific tree structure in ML-MCTDH. More generally, the trees in ML-MCTDH correspond to Tree Tensor Network States (TTNS), which are now commonly used in TD-DMRG. As previously explained, simulations in MQD using ML-MCTDH do not rely solely on the algorithm used to optimize the functions (i.e., the equations of motion derived from the variational principle); they also involve a sophisticated machinery of numerical techniques that have been developed to treat the quantum motion of the nuclei. These include, for example, a variety of primitive basis sets, integrators, and other tools — many of which are also used by the other methods discussed in this review, even those that do not employ ML-MCTDH. Naturally, the same is true for DMRG, where a wide range of different techniques have been developed in various contexts. As H. Larsson concludes, the two methods should not be seen as competing approaches. Rather, there is "much room for mutual cross-fertilization of ideas" between the two communities, which could help broaden their fields of application and improve the underlying techniques. In this context, we note, for example, that H. Larsson and coworkers are using DMRG to calculate vibrational or vibronic eigenstates of various molecules such as the protonated water dimer (seven atoms) [530-534], and P.-N. Roy and coworkers apply DMRG to study the quantum properties of molecular chains [535-537]. In the same manner, MCTDH is being used for electronic dynamics and quantum chemistry calculations [491,493,538-542].

Table 1. Typical maximum number of atoms that can be treated in full dimensionality with methods based on the solution of the Schrödinger equation for the nuclei for different types of processes. Precise examples and the corresponding references are given in the text.

Type of process/quantity	Method	Number of atoms
Rovibrational Eigenstates/high anharmonicity	Time-Independent (TID)	7 (15 DOFs)
Reactive molecular Collisions	Time-Independent (TID)	4 (6 DOFs)
Reactive molecular collisions	Time-Dependent	6 (12 DOFs)
Process with ab initio PES/rectilinear coordinates	ML-MCTDH	25 (75 DOFs)
Process with ab initio PES/curvilinear coordinates	ML-MCTDH	19 (51 DOFs)
Processes described by models	ML-MCTDH or its variants	Hundreds of atoms

To make ideas very clear, we summarize in the **Table 1** the (approximate) maximum number of atoms that can currently be handled using methods based on the Schrödinger equation for different types of calculations. These numbers may change rapidly in some cases, as explained in this review.

5. Discussion and conclusion

By the end of this review, the attentive reader may have realized just how many different numerical methods have been developed—and often combined—to solve the Schrödinger equation for describing molecular systems. He or she may also have recognized that major breakthroughs have been achieved, even very recently. Nevertheless, it is unlikely that a major leap forward will be made in the near future regarding the size of systems that can be treated with very high accuracy, particularly when calculating eigenstates, processes influenced by long-lived quantum resonances, or, more broadly, in the field of ultracold chemistry. Yet, such calculations remain essential, not only for their practical applications, but primarily because they provide crucial benchmarks that are necessary for understanding the quantum behavior of much larger systems.

We can now, nevertheless, be optimistic about the future of MQD. We are at a turning point in this field, where some of the most important methodological developments have been established. The Multi-Layer MCTDH approach and its numerous variants have demonstrated that the treatment of much larger systems is indeed possible. The systematic use of parallelization has, of course, greatly contributed to this progress. The availability of software packages such as the Heidelberg MCTDH package and the QUANTICS package—which includes the former—gives us

reason to hope that it will soon be possible to treat the quantum evolution of larger systems in a much more systematic way. The main focus in the near future will be to use these tools and fully develop their applications to a wide range of systems.

The use of Machine Learning has significantly improved the accessibility of high-quality potential energy surfaces (PESs), particularly for the electronic ground state. It can be anticipated that Machine Learning techniques, and artificial intelligence more broadly, will continue to play a key role in enabling the study of ever larger systems. Ideally, one would like to have access to high-quality potential energy surfaces (PESs)— including those for coupled excited states— directly in a form that is easy to implement, such as a sum-of-product form with a limited number of terms. This is likely a key requirement for drastically extending the precise quantum treatment of very large systems. The possibility that quantum computers may become available in the future makes this prospect not entirely unrealistic.

In conclusion, let us adopt a broader perspective. The methods described here are not intended to address every problem involving nuclear quantum effects in chemistry. Instead, we emphasize their complementarity with more approximate approaches, such as semi-classical methods or path integral techniques. While the latter are exact in principle, their practical utility often lies in combination with approximations, enabling the efficient treatment of extremely large systems. Although the methods discussed in this review can describe condensed phases and incorporate temperature effects, they typically do so under specific conditions. It is reasonable to expect that techniques like Ring Polymer Molecular Dynamics (RPMD) will become more systematically applied, particularly for condensed-phase systems at elevated temperatures. We have highlighted that hybrid methods are already used in MQD: for example, an “active” core may be treated with ML-MCTDH, while the surrounding environment is handled by different, more approximate approaches. Such strategies are likely to play a crucial role in the future, as they allow the integration of methods with distinct advantages and limitations. One could thus envision combining MCTDH not only with path integral methods but also with other “exact” formulations, such as phase space formulations of quantum mechanics [543,544].

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