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Original Article

The 2025 Roadmap to Ultrafast Dynamics: Frontiers of Theoretical and Computational Modelling

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Abstract: *Ultrafast dynamics is among the most revolutionary topics of modern physical science, and it links up photochemistry with materials research and basic quantum study/hot quantum technology. The dynamics of electrons, spins and nuclei that determine the microscopic fundamentals of energy conversion, information transfer and structural change is observable for researchers through ongoing probing (and manipulation) of matter on timescales from femto (10^{-15} s) to attoseconds (10^{-18} s). An analogous revolution in theoretical and computational simulations has been ignited by the unprecedented resolution of the state-of-the-art experimental instruments, such as time-resolved photoelectron spectroscopy [2-5], ultrafast X-ray and electron diffraction [6-8], attosecond laser pulses [9]. These developments demand development of new theories that grasp mechanistic understanding over different scales and be capable to predict the nonequilibrium phenomena efficiently. Here in this article, we provide a comprehensive description of the opportunities opened and challenges at hand in ultrafast theory with the perspective to 2025 within "The 2025 Roadmap to Ultrafast Dynamics: Theory" which complements other roadmaps on Extreme Light Infrastructure (ELI) science as well as on collider and neutrino communities. It discusses the progress and limitation of some major computational approaches, including newly developed quantum-electrodynamical density-functional theory (QEDFT), nonadiabatic molecular dynamics (NAMD), nonequilibrium Green's function (NEGF) methods, and real-time time-dependent density functional theory (TDDFT). The roadmap illustrates how these methods are interfacing with data-driven models and machine learning (ML) and hybrid physics-AI frameworks that accelerate simulations while maintaining physical interpretability.*

Pointed to byLine 300 All these concerns and more are elaborated further on such as the need for reproducible community benchmarks, the potential tradeoff between accuracy and scalability, and connection of ultrafast calculations with experimental observables. The work also lays out several crucial research priorities for 2025–2030, such as the development of open and interoperable software infrastructures, benchmark datasets for nonequilibrium out-of-equilibrium dynamics, or ML-assisted multiscale models capable of describing correlated systems in strong fields. The roadmap also brings out the importance of open-source toolchains, interactive experiment-theory feedback loops that can potentially impact experimental steerage in real-time, and FAIR (Findable, Accessible, Interoperable, Reusable) data principles. This work also provides researchers, educators and politicians who want to develop ultrafast modellings as a predictive and interpretive tool with the first stepstone by describing concrete objectives and the multidisciplinary way forward. Beyond the goal of increasing computer power, the aim is to develop a coherent open sustainable scientific environment that allows for quantitative control of matter on its natural time and size scales. Ultimately, predictive ultrafast theory will enable transformative discoveries in energy materials, photonics, quantum information and catalysis as it will usher a new era of computational research and development grounded on real-time quantum dynamics.

Keywords: *Strong-Field Phenomena, Multiscale Simulation, Benchmarking and FAIR Data, Computational Materials Science, Theoretical Photochemistry, Ultrafast Dynamics, Attosecond Science, Time-Dependent Density Functional Theory (TDDFT), Nonequilibrium Green's Function (NEGF), Nonadiabatic Molecular Dynamics (NAMD), Quantum Electrodynamical DFT (QEDFT), Machine Learning in Physics.*

I. INTRODUCTION

A. Ultrafast Science's Emergence

In the course of the past 20 years, experimental and theoretical abilities to understand the basic dynamics of matter have remarkably joined. A well-established yet fast-growing field at the crossroads of physics, chemistry, and materials science, ultrafast science, i.e. the study of phenomena at timescales of femtosecond (10^{-15} s) and attosecond (10^{-18} s), is based on natural timescales that correspond with the fundamental microscopic mechanisms behind chemical reactions, charge transport, spin interactions, and phase changes: the motion of electrons and nuclei inside atoms and molecules. The course of experiments has been significantly changed by the development of light pulses of dimensions shorter than one oscillation cycle of an optical field. Observations of these dynamics became practicable via these environmental constraints,

such as attosecond spectroscopy, time-resolved photoelectron emission, ultrafast X-ray diffraction, and free-electron lasers. But, to translate these experimental observables into mechanistic insight, there is a need for theoretical frameworks that can explain nonequilibrium processes outside the Born-Oppenheimer approximation and perturbative linear-response theories. fundamentally, ultrafast dynamics is controlled by the time-dependent Schrödinger equation, which is impossible to solve exactly computationally for practical materials. Thus, it is used to achieve feasible hierarchy of approximations between scalability and accuracy. Nonequilibrium Green's function methods, which provide a many-body theory of interconnected systems, and time-dependent density functional theory techniques, which describe the static DFT formalism onto time-dependent location, would be the most habitats. Only when accompanied by molecular dynamics do these techniques allow for the simulation of electron-nuclear interactions and relaxation processes after photoexcitation.

Every approach, however, has its drawbacks. The predictive power of TDDFT is dictated by the choice of the excitonic exchange-correlation functionals and may fail for charge-transfer or strong coupling systems. Despite their formality, NEGF methods often do not scale particularly well to system size and/or duration. While offering a reasonable trade-off, such nonadiabatic molecular dynamics (NAMD) methods that combine quantum electronic transitions and classical nuclear trajectories are still strongly approximate in the system coupling. Thus, hybrid multiscale theories applicable to big realistic systems that can also capture quantum coherence and energy conservation will comprise the 2025 frontier.

B. Big Data, Machine Learning and Data-Driven Discovery

A paradigm shift has taken place since the introduction of machine learning (ML) to ultrafast theory. At the present time, data-driven approaches are employed in the inverse design of laser pulses (via shape function learning) potential-energy surface fitting nonadiabatic-coupling prediction [7] ultrafast spectroscopic signal classification. Purely empirical models, however, have the risk of violating physical laws or not working when extrapolated. Thus a challenge for the next decade will be physics-aware machine learning (ML) with explicit modeling of symmetries, conservation laws, and quantification of uncertainties. By integrating machine learning (ML) and existing ab-initio schemes, such as TDDFT or NEGF, interpretable results can be obtained at an order of magnitude lower computing costs. Also, instantaneous simulation and experiment feedback is possible by using deep learning on experimental data streams from ultrafast electron diffraction (UED) and X-ray free-electron lasers (XFELs). This closed-loop scenario is a crucial aspect of the 2025 roadmap: theoretical predictions guide experiments, which refine theoretical modeling.

C. Novel Physical Domains and Interdisciplinary Cross-Fertilization

Ultrafast processes are inherently interdisciplinary. They elucidate the electron, spin and phonon interplay in condensed matter systems that leads to emergent behaviour such as photo-induced phase transitions, charge density waves or superconductivity. They visualize the quantum wavepacket that accompanies these processes in molecular systems responsible for biological energy transfer and photochemistry. Ultrafast processes enable strong light-matter coupling regimes in quantum optics, where electronic and photonic degrees of freedom mix to form polaritonic states. The new area of QEDFT is a quest to bridge the practice in condensed-matter and chemical theory and in QED, for systems as complex as this. By considering photons and electrons on an equal footing, QEDFT builds upon traditional electronic-structure methods to enable investigation of cavity-managed chemistry and light-induced material phases. This research lays a timely foundation for the next generation of predictive modeling with exascale supercomputing power.

D. The Imperative for A 2025 Strategy

Despite these progress, there remain structural barriers to progress within the ultrafast modelling community including fragmented so``ftware ecosystems, limited use of standardised benchmarks and the relative scarcity of method interoperability and diagnostics or training in experiment for early career researchers. To overcome these challenges, the 2025 Roadmap presents an integrated view taking into account training of skills, data infrastructure as well as methodological advancement.

Important suggestions consist of:

- Open, FAIR-compliant benchmark datasets for nonequilibrium dynamics should be developed to ensure that results are reproducible and transparent.
- Developing interoperable, modular software routines to connect spectroscopic simulations with dynamics solvers and electronic-structure codes.
- With embedded machine learning accelerators to realtime predict and analyze within the limit of physics.
- by fostering co-design environments for experiments and theories where information is exchanged both computational and experiment platforms.
- Promoting sustainability by ensuring equitable access to the shared pools of computational resources, and promoting energy-efficient computing.

E. Impact and Scope of this Paper

This paper provides a strategic plan of the theoretical and computational ultrafast science over the period 2025-2030, along with an interpretational digest of these objectives. It is intended to help promote progress beyond ad-hoc modeling towards a predictive, interoperable, and data-rich ultrafast community environment by drawing upon insights from present-day community reviews, software consortia, and experimental collaborations. The ultimate goal is to connect basic discovery to applied innovation by advancing our understanding of ultrafast processes and the ability to design materials and devices under nonequilibrium conditions. In summary, the science of ultrarapidity stands at a crossroads. Combining machine learning, scalable theory and high-fidelity experiments offers the prospect of moving real-time quantum dynamics from a specialism to design toolkit. This road map prepares that ground by directing the community toward a single framework with which to understand, model and manipulate nature's fastest processes.

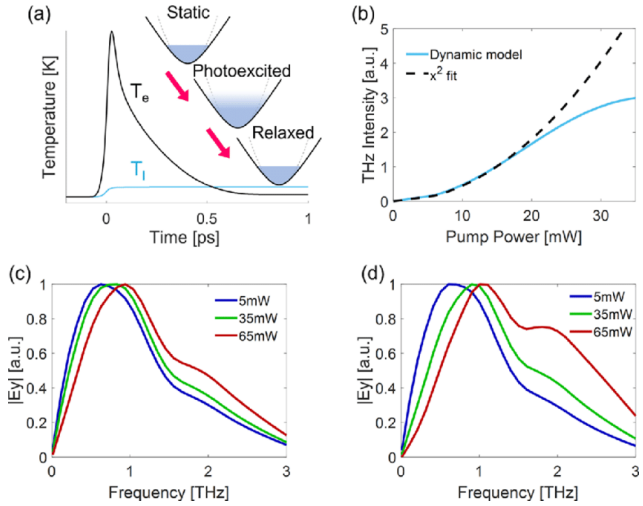

Figure 1 : Schematic of Ultrafast Dynamics and Theoretical Modelling — Non-Adiabatic Transitions, Energy Surfaces, and Time-Resolved Computational Pathways.

II. CONCEPTUAL AND METHODOLOGICAL LANDSCAPE

Ultrafast dynamics aims to describe non-equilibrium matter by a wide range of methods, at both isclussive and computational level. The underlying assumptions, numerical scalability, and physical realism associated with these three methods differ but, as a whole, they represent the building blocks of modern ultrafast theory. The main paradigms are described in the following subsections with emphasis on their successes, current challenges, and frontiers.

A. Time-Dependent Ab-initio Electronic Structure

The fundamental framework to describe the dynamics of electrons in atoms, molecules and solids under exposure to ultrafast fields (td) is provided by Time-Dependent Density Functional Theory (TDDFT). TDDFT is an ideal tool for modeling light-matter interactions and excited-state dynamics because unlike ground-state DFT, it follows the time-dependent response of the electronic density to time-dependent external potentials. New developments, like current-density functional theory (CDFT) or quantum-electrodynamical DFT (QEDFT), to treat and predict polaritonic states in optical cavities as well as light-induced phase transitions involve now matter interacting with quantised radiation fields.

Despite being computationally efficient and intellectually appealing, TDDFT suffers from several limitations. Attosecond and femtosecond timescales are essential for the discussion of long-range charge transfer, multiple excitations and high electron correlation that challenge standard exchange-correlation (XC) functionals. For the hybridization of TDDFT with wave function or Green's function theories at higher level of theory, non-adiabatic and memory-dependent kernels, extensions based on density matrices and embedding methods are under study. Moreover, the hybridization of TDDFT with real-time propagation algorithms and potential energy surfaces built upon machine learning is providing access to scalable yet accurate simulations that bridge first-principles calculations to the very fast time scale of experimental observables.

B. Many–Body Perturbation Theory and NEGF

Our starting point is many-body perturbation theory developed in terms of nonequilibrium Green's function (NEGF). A quantum-statistical theory to describe time-dependent processes in correlated systems is provided by the nonequilibrium Green's function (NEGF) formalism. Using the self-energy functionals and contour-ordered Green's functions, NEGF can also address quantitatively decoherence (including continuum of leads), relaxation, and dissipation by treating electron–electron, electron–phonon, and electron–photon interactions on an equal footing. Transitory photoemission spectra and

nonequilibrium population dynamics are directly accessible via NEGF, which retains correlation and spectral property information, as opposed to TDDFT that only evolves one-body densities.

The two-time propagation costs for NEGF scale quadratically with the time and, thus NP-NEGF is beset by its computational scaling: hardly feasible calculations are possible only for small systems or model Hamiltonians. At the present, active research efforts are devoted towards the development of sampling techniques, scaling laws and embedding schemes which, integrating the efficiency of TDDFT and accuracy of NEGF approaches. For strong laser fields hybrid TDDFT+NEGF or DFT+DMFT approaches give hope for a description of coupled systems. Finally, retaining important correlation effects, reformulations using the generalised Kadanoff–Baym ansatz (GKBA) have allowed for greater efficiency. NEGF is expected to become an important method to bridge experimentally measurable ultrafast observables, e.g., time-resolved photoemission and transient absorption spectra with quantum many-body theory as both exascale computers and tensor-network algorithms progress.

C. NAMD or Nondiabetic Molecular Dynamics

In molecular and condensed matter systems, many ultrafast processes involve non-Born-Oppenheimer nuclear motion (1, 6). By allowing for electronic transitions to occur on nonstationary nuclear trajectories, NAMD techniques provide an essential connection between electronic-structure theory and the motion of nuclei. While the former approach solve electron and nuclei wave equations using, e.g., grid-based techniques (e.g. MCTDH), or its multilayer versions, in the latter trajectory-based methods like FSSH (Fewest Switches Surface Hopping), AIMS (Ab-Initio Multiple Spawning) and mixed quantum-classical Ehrenfest dynamics are used.

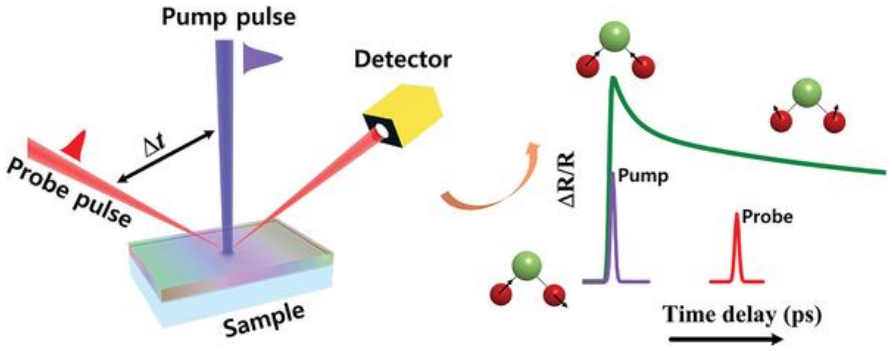

Figure 2 : Computational Simulation of Time-Resolved Ultrafast Phenomena: Modelling of Pump-Probe Processes and Electron/Phonon Dynamics.

The decoherence corrections were recently refined in order to describe more realistic population relaxation, and to obtain smoother transitions between the A states. A second novel development is the machine-learning-accelerated NAMD approach that drastically reduces the computational expense based on employing neural-network potentials to mimic ab-initio PESs with near chemical accuracy. Nevertheless, the formulation of nonadiabatic couplings as well as the used electronic-structure method have a large impact on the quality of NAMD predictions. Therefore, the community is now moving towards protocols for uncertainty quantification and cross-method validation together with standardised benchmark datasets. To achieve predictive modelling of photochemical reactions, charge transfer, and structural switching in realistic condensed-phase and biological environments, the next steps will integrate on-the-fly quantum chemistry, ML-based PESs and statistical sampling methods.

D. Theoretical Interlude Photoionisation and Attosecond Electron Dynamics

Real-time observation of electron motion can now be carried out using attosecond spectroscopy, in which charge migration, tunnelling ionisation and photoemission time delays have all been investigated. For theoretical modelling in this regime, the many-body and nonperturbative nature inherent to multi-electron systems under intense laser fields demands solution of the time-dependent Schrödinger equation. To account for the interplay of correlation, spin–orbit coupling and continuum dynamics in modern approaches MC-TDHF/TDDFT 11, time-dependent R-matrix (TDRM) 12, strong-field approximation (SFA) 10 are used. The accurate calculation of photoelectron circular dichroism (PECD) and PADs, which carry information on electronic phase, chirality, and coherence provides a major achievement in this domain. Moreover, interference or attosecond streaking measurements of angle-resolved time delays can deliver stringent tests of theoretical accuracy. The current challenge is to correlate such observables with 'instantaneous' electron correlation in time, using light,details on this and logical control strategies that influence charge transfer with custom light beams are at the present frontier. Once resources grow enough to allow real-time interpretation of experimental signals and a deeper insight into the quantum choreography of electrons in complex systems, attosecond theory gets increasingly intertwined with ML-driven data analysis and reduced-order models.

E. Imaging and Ultrafast Scattering (XFEL, UED)

Imaging in the broad sense includes ultrafast scattering of X-ray free-electron-laser(XFEL), ultrafast electron diffraction(UED), etc.and imaging diagnostics can be applied in these cases. Ultrafast electron diffraction (UED) and time-resolved X-ray free-electron laser (XFEL) sources provide atomic spacetime resolution for images of material structures, with both femtosecond and atomic precision. Theoretical modeling of such experiments is necessary to translate diffraction patterns into dynamic structural and electronic information. Coupling between nonthermal lattice-vibration, transient charge redistribution, and laser-induced electronic dynamics is characterized by means of an elaborate extension of traditional diffraction theory. Quantitative descriptions concerning time-dependent structures and the intensities of scattering can now be calculated with ab-initio electron-nuclear simulations and compared to data.

Combining compressed sensing techniques with deep learning-based phase retrieval to reconstruct real-space charge densities from noisy or incomplete diffraction data is one of the latest advancements in this area. These processes offer real-time response during experiments and greatly accelerate data inversion. In principle multiscale simulations which also involve TDDFT, classical dynamics and real-space diffraction modeling are required to bridge scattering observables with the electronic structure(f8,16,f20). Future work involves data-driven uncertainty quantification, coherent diffraction imaging of electronic currents and modeling of pump-probe sequences. Taken together, these advances are making ultrafast scattering a quantitative tool for probing the structural and electronic dynamics of molecules, nanoparticles, and the condensed phase.

F. Hybrid Physics-ML Models and Machine Learning (ML) models

The field of ultrafast dynamics modeling has seen a major push due to the advent of machine learning (ML), which offers data-driven surrogates that accelerate simulations by orders of magnitude. Nowadays, we use ML algorithms to analyze ultrafast spectroscopic data, fit potential-energy surfaces (PESs), search for reaction coordinates, and obtain nonadiabatic coupling elements. In particular, systems that were previously computationally infeasible due to the immense computational expense can now be treated with near-ab-initio accuracy using neural-network potentials and Gaussian-process regression. However purely empirical model has no foundations in basic principles, such as causality, symmetry or energy conservation that might lead to some non-physical results.

To tackle these challenges, researchers are developing hybrid physics-ML frameworks in which physical rules are embedded directly into the model design. These consist of energy-conserving recurrent networks ensuring long-term dynamical stability, symmetry-equivariant models over graphs, and Hamiltonian neural networks. Integrating machine learning (ML) with Bayesian inference and uncertainty quantification is a further frontier allowing adaptive sampling and error estimation in simulations. In the laboratory, ML aids in inverse design of laser pulses for coherent control and accelerates the analysis of large datasets from XFEL and UED sources. Last but not least, by porting the scalability and flexibility of data-driven methods to first-principles simulations these integration frameworks herald a new era in ultrafast simulation towards autonomous discovery and real-time predictive simulation.

III. KEY BOTTLENECKS AND CHALLENGES

Despite revolutionary breakthroughs of theoretical and computational techniques, the predictive power, scalability, interoperability remain to be seriously constrained by both fundamental issues and practical problems in ultrafast dynamics. Time-resolved experiments and models of complex materials are pushing this trade-off between computational tractability and physical rigour to the limit. The current major challenges that now dominate the ultrafast research and modeling frontier are reported in this section.

A. Tradeoffs in Accuracy and Scalability

The challenge of scalability versus exactness is one of the oldest ultrafast problems. Realistic systems such as biological chromophores, hybrid perovskites or nano-structred catalysts that contain hundreds to thousands of atoms indeed induce strong couplings between nuclear and electronic degrees of freedom. High-accuracy methods such as nonequilibrium Green's functions (NEGF), coupledc̈cluster (CC) and configuration interaction (CI) exist to provide accurate physical resolutions,5 6 but have exponential or quartic compute complexity with system size such that they become immediately unsuitable for long systems.

On the other hand, for more scalable methods, such as (semi)empirical tight-binding methods or TDDFT, accuracy is compromised especially in the case of highly coupled or multi-excitonic events. Accordingly, the community is actively developing local orbital crystal-field-like orbitals,b reduced-scaling many-body perturbation theories,c and embedding schemes to hybridize dierent theoretical levels of theory.d Herewith, we present a wide-range benchmarking for the relatively new exciting code with respect to electronic structure methods as well as experimentation. By operating at a coarser level of description, these multicycle methods (such as DFT-in-GW or TDDFT-in-CC) allow computational efforts to

be focused in areas where a system is most active. Simultaneously, ML surrogate models have emerged as promising accelerators which achieve near ab-initio accuracy at a fraction of the cost. It remains challenging, however, to ensure the physical consistency and transferability of these models. It aims to construct a computationally feasible, scalable and systematically improvable theoretical approach away from the integrability limit which retains basic many-body physics for systems of thousands of degrees of freedom.

B. Comparison and Replicability

Unified benchmarks and repeatability standards are urgently in demand in the field, as ultrafast simulations become increasingly common on materials that cover a broad range of timescales. Despite the overall encouraging progress, it remains difficult to compare directly results obtained from different research groups (most of them have used also different software implementations), approximations or initial conditions. This level of incompatibility, therefore impairs any evaluation of methods on a neutral basis and impedes the uptake of "best practice" agreement.

Since time-dependent observables such as population transfer, decay rates or photoemission delays depend on integration methods, basis sets and possibly gauge options, benchmarking of nonequilibrium dynamics poses unique obstacles. Their nonequilibrium analogues are still in their infancy, while equilibrium electronic-structure benchmarks (e.g., GMTKN55 for thermochemistry, or the MLatom datasets for molecular dynamics) have become community standards. The present roadmapping activities in photochemistry and in ultrafast theory aim at establishment of open- access benchmark databases involving information on condensed phases, molecules and clusters obtained under various excitation conditions. These data sets need to include comprehensive methodological metadata (versions, functionals, convergence parameters, and input geometries) as well as simulation results to ensure reproducibility. Replicable computational ultrafast science is increasingly being enabled by collaborative platforms that allow process automation and cross-code validation (as in the case of AiiDA or Atomic Simulation Environment).

Ultimately, reproducibility is not simply a technical issue but also an enabling demand for the inter-operation of theory-based simulation and experiment as well as scientific authority.

C. Multi-Scale Coupling and Time Scales

Ultrafast experiments are naturally sensitive to multiscale dynamics because electronic, vibrational and spin degrees of freedom evolve on separate but related time scales. For example the spin pattern can persist for nanoseconds, a lattice can respond in picoseconds and an electron can tunnel in attoseconds. To simulate these coupled phenomena, a unified theoretical framework capable of relating quantum electronic dynamics to mesoscopic or even macroscopic observables is required.

Most existing methods artificially partition the the domains but specializing to either an entirely quantum mechanical or semi-classical description as the atomic and electronic scales are integrated. It is still challenging to devise multi-scale coupling schemes that are able to automatically switch among theoretical levels. Coarse grained quantum embedding76,77 and hybrid Ehrenfest to Langevin techniques78,79 and similar forms of quantum classical Liouville methods80 are new strategies aimed at interfacing electronic excitations with thermal environment. Another frontier is the real-time coupling of quantum and continuum degrees of freedom which is essential for connecting microscopic dynamics to experimental observables such as pump-probe diffraction patterns or transient optical spectra. Progress in that direction would enable experimentally interpretable and time-resolved simulations of spin, lattice, as well as electronic sub-systems into a single pre-dictive model.

D. Data Load and Instantaneous Experimental Feedback

Modern ultrafast experiments, such as highharmonic generation (HHG),3–5 ultrashort electron diffraction1 (UED) and X-ray free-electron lasers7,8 (XFELs), generate data at rates that previously could not be imagined, often exceeding tera-bytes of data per experiment. Simply making the best use of massive volumes of data is a huge problem. New detector technologies with high throughput are no longer compatible with traditional processing pipeline algorithms which depend on offline postanalysis. To tackle this, academics are building low-latency algorithms, streaming data frameworks and machine-learning aided feedback loops that can analyse experimental results in real time. These systems adopt probabilistic models and physics-informed neural networks that are trained on synthetic data, originating from simulations, to search for anomalies, learn trends, and dynamically guide experimental interventions. Another challenge is to maintain the physical interpretability of ML-oriented analysis tools. Unless constrained by physical priors, these are nothing but purely statistical correlations and may obscure causal processes. As a result, physics-aware AI with training on conservation laws, symmetry operations and known spectral signatures, profound in the emerging data-driven ultrafast science. This combination offers the prospect of a new level of closed-loop ultrafast experiment, in which theory, computing and experiment co-evolve in real time.

E. Materials Modelling together with Quantum Optics

Quantum-optically driven, materials modelling is a rapidly expanding but conceptually demanding aspect of the ultrafast science. The quantisation of the electromagnetic field and electronic states must necessarily be considered together because strong light-matter interactions and polaritonic chemistry have become important. These effects are not fully accounted for in standard materials theories that rely on a classical or semi-classical description of the electromagnetic field and quantum-optical models systematically neglect atomistic features necessary for quantitatively accurate predictions.

Conceptually, recently developed polaritonic many-body perturbation theory as well as quantum-electrodynamical density functional theory (QEDFT) give rise to a theoretical formalism for describing such coupled photon–electron–phonon systems. These methods remain computationally intensive and have not been yet standardized for use with various material platforms. The main challenge is extending these features to useful solid-state environments including dissipation, cavity modes, as well as nonlocal screening. Besides algorithmic efficiency, validation against benchmark experimental observables such as cavity-induced reaction rates, Rabi splittings, and photonic band shifts are required for broader application. After achieving this synthesis, we hope to predict the design of quantum materials in optical cavities, with exciting prospects for polaritonic transport, light-driven superconductivity and coherent control of inter-electron correlations.

F. Summary

The future of science in 2025 and beyond, when it comes to ultrafast science, can be summed up based on the four challenges just outlined: data management, quantum optics integration, scale bridging, reproducibility as well as accuracy versus scalability balance. Culturally, an open, interoperable and interdisciplinary way of working linking theory, computation, and experiment in a tight feedback loop is as key to solving them as methodological novelty.

IV. ROADMAP: PRIORITIES, MILESTONES, AND RECOMMENDED ACTIONS (2025–2030)

The next five years will be the key period required to take ultrafast theoretical and computational research away from a set of specialised methods towards an integrated, interoperable field that is capable to predict models across materials and timescales. This roadmap, involving the challenges mentioned previously, transforms strategic priorities into technical actions that address scalability, reproducibility and real-time theory/experiment collaboration. On a five-year time scale, the establishment of a large number of community benchmark issues including molecules, low-dimensional systems, and bulk solids is our highest priority. Standardized input geometries, simulation sequences and experimentally validated observables such as time-resolved spectra, photoelectron angular distributions and ultrafast diffraction pattern are identified to be featured by these benchmarks. These benchmarks also will serve as reference points for testing reproducibility and validating across methods, for example, because they supply canonical test sets and standardized output formats. The open publication of at least twenty benchmark systems, along with many independent method comparisons, can be used to measure how this shared task is doing and to set up a solid framework for assessing new hybrid approaches.

In addition, the community needs to converge on open and compatible software toolchains by 2025-27. Lack of integration between molecular dynamics engines, analysis frameworks and electronic structure codes further cause that efforts are being duplicated and methods are hard to be interoperable. Modular libraries and standardised data formats, plus shared APIs (such as application programming interfaces), would enable seamless data exchange between phenomenological propagators based on machine learning techniques, TDDFT, NEGF etc. For longevity beyond individual groups, however, such infrastructure needs to foster collaborative development under community governance models. An appreciation of success would be to encourage use of an uniform input/output schema by important software packages, accompanied with worked reference workflows that combine spectroscopic calculations, nuclear and electronic structure simulations in one single pipeline. Physics-informed machine learning (ML) is expected to profoundly change the ultrafast-modelling community in 2025-2028. The focus should shift from purely data-driven models to hybrid designs which both quantify the forecast uncertainty and encode conservation laws, like charge and energy conservation. In order to ensure applicability across regimes, these models will have to be trained on both synthetic (simulation-based) and experimental datasets. An important benchmark for performance will be the ability to use ML surrogates that can facilitate at least an order of magnitude speed-up in computations while maintaining errors below five percent on benchmark observables and with uncertainty estimates consistent with physical expectation. These techniques have the potential to provide real-time simulation of light-driven processes, something that was not previously feasible.

An analogous priority exists in the mid-term (from 2026 to 2029) for the development of scalable many-body approximations and reduced representations. In order to push simulations of correlated electrons from toy model systems to real materials new numerical algorithms, which are able to treat very large active regions and long simulation times without getting unaffordable, have to be developed. Promising methodologies include time-domain versions of dynamical mean-field theory (DMFT), reduced-scaling NEGF solvers, and inexpensive self-energy approximations. The scalability can be further

enhanced by incorporating the frameworks that link less approximate environments to high-accuracy correlated regions. Quantitative comparison between, e.g. transient spectral shifts or coherence rates with time-resolved experiments and the ab-initio simulation of correlated ultrafast dynamics in systems containing more than 100 atoms will be the benchmarks for success. The co-design of experiments and theory with the support of adaptive control and real-time data feedback is another key priority. With increasing amounts of data produced in high-repetition-rate or large-scale facilities such as XFELs and ultrafast electron diffraction (UED) beamlines, it is essential to include low-latency algorithms and AI-based control systems. Real-time analysis pipelines can take detector outputs, fit provisional models, and suggest how the experiment should be modified eg probe-delay tuning or pulse-shape optimisation. Simulations and real experiments will also enable a discovery through closed loop frameworks providing autonomous experimental steering, that optimize beamline efficiency. Within the next 1–2 years the community is expected to reach an important milestone if operation of a functional feedback loop was shown for single photon imaging at a major XFEL or UED facility, showing that ultrafast research can transition from analysis after-the-fact to predictive and self-correcting exploration.

Finally, long-term improvement needs funding for workforce development, education and open community practices. Because the field is multidisciplinary, researchers ought to be able to speak both the language of computation and experiment — they should be capable of using simulations based on experiment or experiments informed by theory. This bridge will be supported by the promotion of open coding workshops, summer schools and cross-disciplinary training toolkit. Results, procedures and metadata will be also saved for future validation and reuse in case the ultra fast dynamics datasets are shared adhering to FAIR data principles (Findable, Accessible, Interoperable, Reusable). Within two years we want to see leading journals in the domain regularly publish datasets as FAIR-compliant when taking submissions, and teach at least 100 researchers through structured workshops. These combined efforts result in a logical vision for the road map of 2025–2030 including setting trust stand-ards, tooling for openness in teamwork, ML acceleration of processes and methods, realism through many-body methodologies scaling beyond DFT, theory meets experiment to increase agility and education towards sustaina-bility with modelling. By linking milestones to measurable outcomes, the ultrafast community could move from a fractured process of methodological development to an integrated ecosystem able to predict and control matter on the fastest time scales known in science.

V. EXAMPLE USE-CASES AND ILLUSTRATIVE WORKFLOWS

In this section, we present three sample workflows for the implementation of theory, computation and experiment in ultrafast research to ground the road map in a specific scientific approach. These benchmark examples illustrate how physics- aware machine learning (ML), experiment-theory feedback loops, open software-frameworks, and community-recommended benchmarks can together facilitate progress in real-time discovery and predictive modeling.

The first example involves one of the most clear type of electronic cogernce in chemistry -- attosecond charge migration in organic molecules. This work begin by developing accurate ground and excited state electronic structures using hybrid GW + Bethe–Salpeter equation (BSE) or constrained time-dependent density functional theory (TDDFT) methods. These provide the relevant correlated transition densities and excitation energies needed to specify coherent electronic state superpositions driven by attosecond pulses. The resulting electron wavepacket is then time propagated adiabatically in the presence of external laser fields. Here the backbone is formed by real-time TDDFT which can be partially corrected with NEGF self-energies, to describe relaxation effects and electron-electron scattering not included in standard TDDFT functionals.

Time-dependent charge densities and dipole moments resulting from the simulated dynamics are directly compared with time-resolved photoelectron angular distributions (PADs) measured in attosecond pump–probe experiments. Model parameters such as the laser intensity, pulse phase and exchange-correlation kernel are varied through Bayesian inversion techniques and uncertainty quantification to achieve quantitative agreement. Reliable error estimation and vastly faster convergence can be obtained by training machine-learning surrogates on inexpensive TDDFT trajectories, which enable orders-of-magnitude more sampling of molecular conformations and laser configurations. The end product of this - collection of PECD maps and PADs combines appropriate shape of calculated charge motion with electron symmetry in the experiment. The present application demonstrates an example of how predictive modelling can inform attosecond spectroscopy mechanistic interpretation and pulse design.

Second, an example workflow studies the ultrafast switching in correlated materials relevant to data-storage and next-generation quantum devices. In particular, simulations based on nonequilibrium Green's functions (NEGF) or time-dependent dynamical mean-field theory (DMFT) are the theoretical framework here. These frameworks can account for how ephemeral phase transitions, such as insulator-metal switching or the melting of charge-density waves, are facilitated by a strong interplay of electron-electron interactions and light-matter coupling. Order-parameter time traces, such as magnetization, polarization or gap size are part of the simulated output under specialized laser excitation. By comparing

these calculated response functions with the time-resolved optical conductivity and X-ray diffraction, a one-to-one map is obtained between theory and experiment.

Closed-loop co-design with its experimental partners is one key part of this approach. A target dynamic end result A goal of the kind can be realized by feedback control to the numerical tools using machine learning (ML)-based optimisation that repeatedly adjusts laser parameters, including intensity, polarisation, pulse sequence or/and wavelength, within a simulation–experiment loop. The approach is illustrative of how theoretical models, computational pipelines and experimental control can create an adaptive feedback loop that transforms discovery from a pure find-it-by-trial-and-error to one where results are obtained in an evidence-guided manner. The ultimate goal would be routine real-time predictive control in the presence of intense laser excitation of correlated materials. The third example considers time-resolved diffraction and imaging at the meso- and nanoscales where structural and electronic degrees of freedom co-evolve. This is a dynamical method where atomic motion from Ehrenfest or nonadiabatic molecular dynamics (NAMD) trajectories is used in conjunction with electronic density evolution from TDDFT or NEGF simulations. For the generation of simulated UED or XFEL images, this composite dataset provides actual structural arrangements and electronic charge distributions which can be forward propagated. These forward simulations are critical for the interpretation of experimental diffraction data, which is typically noisy and has incomplete phase information.

Machine-learning-based phase-retrieval methods accelerate the inversion process – that is, from diffraction patterns to real-space time-dependent structures. Such models give in situ feedback during the experiment, as they are able to reconstruct instantaneous charge and lattice distributions nearly in real time from training on large ensembles of simulated images. This pipeline, from atomic-scale dynamics to experimentally observable observables is comprised of high-fidelity theory, scalable computation and inverse from data. This approach show the possibilities that first-principles modelling and measurements of ultrafast scattering can provide for never-before-seen details on the coupled electrical and structural dynamics in materials, nanostructures, and molecular assemblies. Assembled, these three processes demonstrate the sort of inter-play and collaboration anticipated by the strategy 2025–2030. Each of them highlight the transition from closed simulators to open, feedback-driven and interoperable frameworks where experimental data, machine learning accelerators and physical models reinforce each other. From first principles to live analysis of data in 3D, the ultrafast community moves towards a fully predictive and adaptive paradigm, embedding reproducibility, scalability and physical interpretability at each step.

Table 1 : Representative Workflows for Ultrafast Science (2025–2030)

Use Case	Scientific Focus	Core Theoretical Tools	Computational Components	Experimental Integration	Expected Outcome / Impact
Attosecond charge migration in organic molecules	Real-time electron coherence and photoionization	Constrained TDDFT, GW+BSE, NEGF corrections	Real-time propagation, ML surrogates, Bayesian inversion	Comparison with attosecond PADs & PECD; uncertainty-guided model tuning	Mechanistic insight into ultrafast electron motion and light-field control of charge migration
Ultrafast switching in correlated materials	Light-induced phase transitions and correlated order dynamics	Nonequilibrium Green's functions, time-dependent DMFT	ML-driven closed-loop optimization, scalable solvers	Pump–probe optical/X-ray correlation; adaptive laser control	Predictive design of optically switchable materials for ultrafast electronics
Time-resolved diffraction and imaging	Structural–electronic coupling at mesoscale/nanoscale	TDDFT + Ehrenfest/NAMD coupling	Forward diffraction simulation, ML-based phase retrieval	Real-time XFEL/UED data inversion and feedback	Quantitative reconstruction of transient atomic and charge structures in real time

The priorities of the roadmap — community targets, open source software, physics-aware machine learning methods, scalable many-body solvers and real-time experiment-theory integration with significant training in interoperable workflows — are operationalised through these use cases collectively. It is expected that by the year 2030 they will evolve into self-sufficient, data-driven labs capable of predicting, controlling and simulating ultrafast processes on the molecular-to-macroscopic scale.

VI. DATA & SOFTWARE INFRASTRUCTURE: FAIR, REPRODUCIBLE, AND OPEN

The effective execution of the ultrafast science roadmap requires a strong, open and interoperable data and software environment. As experimental and computational methods continue to produce vast, multidimensional data sets from diffraction images to time-dependent electronic densities, the community needs to adopt open, standardised infrastructure underpinning the Findable, Accessible, Interoperable and Reusable (FAIR) organisation of the data produced28. Reproducibility, benchmarking and cross-disciplinary integration will remain fragmented and inefficient in the absence of such an ecosystem. The core of this transition lies in the creation of giga- and terabyte-sized datasets, compliant to the FAIR standards, able to record ultrafast trajectories with sufficiently high temporal, spatial and contextual resolution. These files should include all necessary experimental and computational metadata that is required for reconstitution and analysis of the raw observables – e.g. electron concentrations, charge currents, state populations, or transient spectra. This involves a complete description of all relevant parameters such as pump and probe photon energies, pulse shapes and durations, detection geometry, field strengths, sampling rates or uncertainty models via metadata. By defining the metadata formats across codes and facilities, data from many sources can thus be integrated into coherent analyses and machine learning pipelines; data analysis places that follow these standards are guaranteed compatibility, consistency.

Developed and containerized computational workflows are also of equal importance. With these workflows, which are packaged using open technologies such as Docker, Singularity and Jupyter/Nextflow users will have the ability to reproduce the published figures directly from raw input data. Software dependencies, numerical parameters, and data-processing methodologies also need to be well-defined in each workflow to ensure a fully transparent chain from setup of the simulation to generation of the figure. This example of containerised reproducibility underpins auditable and repeatable research amongst institutions, as well as democratising access to high-end computation. To achieve a verifiable connection between data, code and conclusions journals and funding bodies should probably begin to insist that key statistics in articles are tied to these containerised workflows. All public datasets, workflows and machine learning models should be assigned persistent digital object identifiers (DOIs) to facilitate discovery and promote long-term preservation. These DOIs would provide a preserved reference for future generations of researchers to be able to access the data, while also ensuring that the appropriate acknowledgment and credit is given to those who created the data. To host datasets in a standardized format, domain specific portals are encouraged to collaborate with data repositories like Zenodo, Materials Cloud or institutional repositories. Moreover, it will be possible to carry out large-scale benchmarking and cross-validation -based on simulation-experiment well-integrated datasets with common metadata structures-which will accelerate the convergence of methodologies.

Open, interoperable software toolchains is another significant aspect of the infrastructure project. The community should unite to adopt a common compatible input/output (I/O) and application programming interface (APIs) pipeline for interfacing among tools of data analysis, nonequilibrium dynamics solvers, and electronic-structure codes. Through the realization of modular, interoperable libraries instead of separate monolithic codes, the scientist can combine techniques such time-dependent density functional theory (TDDFT), nonequilibrium Green's function (NEGF) solvers or molecular dynamics engines within unified workflows. Open governance and community-maintained codebases will be important to ensure sustainability, long-term maintenance and openness in algorithm design. Finally, training and community activities that allows researcher to make efficient use of open data and repeatable software infrastructures should be part of such projects. Workshops, hackathons, and cross-institutional training initiatives should primarily focus on workflows for cloud-based data sharing and version control, workflow automation, as well as adoption of FAIR data principles. Inculcating these principles early into the research process will create a generation of scientists for whom open, reproducible science is simply the way that research is done rather than the exception.

Summary To achieve the vision of a predictive and collaborative ultrafast science we need to develop an ecosystem that enforces FAIR data principles and containerisation for reproducibility, long lived IDs, protocols for data exchange, open source Toolchains that can communicate with each other and actively grow session data capacity and provide inclusive Community Training. By adopting these practices from 2025 to 2030, the field will ensure that data, models and findings remain transparent, reusable, and verifiable, transforming ultrafast science into a genuinely open and reproducible discipline.

VII. CONCLUSION

Ultrafast dynamics is one of the most challenging domains in present day physics and computer science. The key paths along which theory, computation and experiment will need to progress to achieve predictive control over matter on the femtosecond (fs) and attosecond (as) timescales are set out in this 2025 roadmap. Ultrafast dynamics represent an unprecedented window on the interaction of electrons, nuclei and photons under extreme nonequilibrium conditions, as boundaries between chemistry, physics and materials science further dissolve. Yet a coordinated international effort that

includes methodological invention, high-performance computing infrastructure, and open scientific working groups is needed to deliver truly multiscale, reproducible and empirically constrained models. The roadmap shows that progress in nonadiabatic molecular dynamics (NAMD), nonequilibrium Green's function (NEGF) theory, and ab-initio time-dependent electronic structure will also be essential to understanding ultrafast electron–nuclear coupling. Combining these with QEDFT and ML, simulations of well-dressed coherent light-matter systems that were previously unfeasible will be within reach. Unified, modular workflows that connect ML-powered surrogates to TDDFT and NEGF will be a key step in this direction—reducing the cost of simulation while preserving its accuracy.

Other cross-cutting problems that the field needs to solve are creating benchmark datasets, scalable many-body approximations and open software infrastructures with suitable compatibility and repeatability. In order to enable transparency, reproducibility and crossinstitutional data reuse, containerized workflows as well long persistent DOIs, FAIR data policy are critical. Novel physics-aware machine learning (ML) methods at X-ray free-electron laser (XFEL) and ultrafast electron diffraction (UED) facilities will also accelerat3e simulations without losing interpretability, thus allowing for real-time feedback loops between theory and experiment. This roadmap also emphasizes the synergy between theory and experiment in closed-loop discovery paradigms as a transformative theme. By integrating low-latency data pipelines with adaptive pulse shaping and Bayesian optimisation, the community can progress towards autonomous experimentation where experiments are designed directly based on theoretical predictions. This synergy will lead to the discovery of new emergent phenomena in correlated materia ls a nd molec ular systems and more rapid decoding of complex data. In 2030 the wealth of communities will depend on sustained investments in open infrastructure, cooperative governance and education. A new school of (computational-experimentalist) researchers will emerge by setting up common benchmarks, data bases and teaching platforms. Ultimately, the ambition of the roadmap reaches far beyond disciplinary boundaries in seeking to establish an ultrafast research ecosystem that is transparent, globally networked and aligned with FAIR principles capable of anticipating and driving quantum matter at its fundamental temporal limits.

VIII. REFERENCES

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