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Editorial

Ultrafast phenomena from attosecond to picosecond timescales: theory and experiments

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1 Introduction

Ultrafast science aims to study and control out-of-equilibrium properties of matter at microscopic length scales. With impressive developments, ultrafast technologies allow the investigation of particle and quasi-particle dynamics, energy flux, as well as structural and phase transformations with picosecond, femtosecond, or even attosecond resolution. Initially confined to model systems and ultraviolet (UV)–visible–infrared (IR) radiation, the ultrafast community has grown considerably and pushed the frontiers of understanding of the inner workings of matter, investigating increasingly complex objects (biomolecules and quantum materials), supported by the development of new, high-performance ultrafast probes (X-rays, attosecond pulses, THz effects, electron beams, and ultra-intense femtosecond pulses). Combined with indispensable theoretical approaches describing light–matter interaction and particle couplings on multiple time and length scales, this research field has created new links between communities from fundamental physics (to study quantum electrodynamics (QED), quantum entanglement, or coherence) to chemistry (catalysis), biology (photoactive proteins), or materials science (structuration, phase transitions).

This special issue is dedicated to the research activity of the French scientific network on ultrafast phenomena (GDR UP no. 3754). GDR UP gathers all the French teams interested in ultrafast phenomena, in all phases of matter, and covers both experimental and theoretical aspects. It is composed of more than 50 laboratories with more than 800 members and is supported by the Centre National de la Recherche Scientifique (CNRS). The GDR UP was created in 2016 for a 5-year period and was renewed in 2021 for five more years. The GDR UP activities include networking through the organization of annual national meetings, workshops, and periodic web seminars. Furthermore, it provides financial support for national collaborations. It contributes to the training of students and researchers though summer schools and to the promotion of this scientific community with, e.g. this special issue in *EPJST*.

GDR UP is structured into five scientific topics (attosecond science, gas-phase systems from atoms to large molecules, femtochemistry and femtobiology in condensed phase, ultrafast dynamics in materials and nanostructures, and secondary sources: electrons, protons, and photons). This special issue provides an overview of current activities in French laboratories, including international collaborations, in all these topics. It thus summarizes recent progress in this field and points out research directions that should lead to significant scientific and technological advances in the coming years in ultrafast science.

We acknowledge the various laboratories (ILM, ISMO, LIDYL, LOA, LOB, LP3, LPS, LCF, IPCMS, IPR, LSI, ICP, LPMS, CEMES, LASIRE, IJCL, LaHC, IMPMC, SOLEIL, LPT, Femto-ST, CEISAM, ICGM, Institut Lavoisier de Versailles, PPSM, LCS, Institut Jean Lamour, IMN, Institut Fresnel, LCT, CEA-DIF-Arpajon, LMCE, and IRMAR) whose contributions have led to the 27 articles presented in this special issue.

2 Attosecond science

The attosecond timescale is the scale of the classical rotational period of electrons around atoms. Having access to the attosecond time scale offers a new means to study electron dynamics on the angstrom length scale. In recent years, the landscape where electrons can be monitored evolved from the one issued from simple isolated atoms to that of complex isolated molecules or the condensed phase. The progress in this field has

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benefited from the development of light sources providing ultrashort pulse radiation at increasingly higher photon energies (VUV–XUV–X-ray) or with topological control. These are combined with new spectroscopic approaches and light characterization tools. This is illustrated in some of the articles in this special issue. In ref. [1], the authors studied attosecond time delays in photoionization of polyatomic molecules using angle-resolved electron spectroscopy and showed how this ionization delay is affected by resonances in diatomic (N_2) and polyatomic molecules (C_2H_4), as well as the possibility of accessing the attosecond dynamics in two-dimensional (2D) molecules (naphthalene) or in the plasmon resonance of C_{60}. Reference [2] describes the development of state-of-the-art versatile beamlines operating at 10 kHz, providing tunable XUV radiation (10–100 eV) with ultrashort pulses (30–0.3 fs) that allows the study of condensed matter using angle-resolved photoelectron spectroscopy (ARPES) or gas-phase systems using coincidence spectrometry. In the context of attosecond physics, the study of small quantum systems requires the development of sophisticated spectroscopy techniques that allow for a complete reconstruction of the dynamics, potentially in the molecular frame. Investigations in this direction are presented in Ref. [3], where electron–ion coincidence three-dimensional (3D) momentum spectroscopy is used to study the dynamics in XUV and XUV–IR configurations. Beyond shorter duration and tunability, the development of XUV light sources with controlled topological properties offers new perspectives to study materials with nanometric and sub-femtosecond resolution. In ref. [4], the authors studied how vector, vortex, and vector–vortex near-infrared beams are converted into XUV radiation. This allows one to tailor the spin and orbital angular momenta of the XUV pulse, providing vector beams with various spatially dependent polarization distributions. High-harmonic generation (HHG) can also occur in solid or plasma, offering promise as a source of intense XUV light sources, yet facing new challenges in terms of light characterization. In ref. [5], high-order harmonics generated from plasma mirrors are studied with the aim of characterizing the radiation at the attosecond timescale using angle-resolved photoelectron spectroscopy.

3 Gas-phase systems from atoms to large molecules

Studies performed in the gas phase provide intrinsic properties of molecular systems. For this reason, they are of strong interest for refining the understanding of dynamics and for benchmarking of theoretical calculations. Time-resolved experiments provide a means to access coupled electron–nuclear dynamics, which also requires the development of accurate calculations. Interactions in extreme conditions, such as high-photon-energy pulses and intense laser pulses, offer new methods to manipulate processes at the quantum level. A the same time, new methodologies offer the opportunity to study more complex systems, in connection with applications.

In ref. [6], the author provides a review of recent theoretical works performed in French laboratories, to describe nuclear and nonadiabatic ultrafast dynamics in isolated molecules. The review focuses on recent studies on quantum dynamics of small molecules or Hamiltonian models, and "on-the-fly" mixed quantum–classical dynamics of molecules. Beyond the weak-field configuration encountered in pump–probe experiments, the investigation of interactions between molecules with intense light pulses, in the IR up to the XUV, also provides exciting examples of ultrafast control. In ref. [7], single ionization of H_2 molecules exposed to strong and short laser pulses is investigated by using a semi-classical method in various intensity, wavelength, and light polarization regimes. The authors found a non-Franck–Condon vibrational distribution in the cation, in both linear and circular polarizations. In ref. [8], hole dynamics in molecules following irradiation with a short intense XUV pulse is computed and reveals unexpected system-dependent instabilities of light-induced time-dependent dipoles in diatomic and atomic clusters. Beyond the investigation of "small" model systems, more complex macromolecules offer a new playground and challenges for theorists, with important potential applications. Reference [9] investigates relaxation dynamics in complex two-dimensional (phenylene ethynylene) oligomers and identifies an unexpected ultrafast relaxation pathway involving in-plane *trans*-bending motions and involving a dark transient state.

4 Femtochemistry and femtobiology in condensed phase

Condensed-phase studies are the in situ and in operando natural environment for photobiology and organic photochemistry processes. This is connected to applications and emerging studies involving condensed-phase femtochemisty and photon energy conversion, such as organic photovoltaic devices and photocatalytic cells. The target is complex systems embedded in an environment, whose elucidation could necessitate the combination of multiple spectroscopic techniques over a wide wavelength range as it implies multiple time/space scales involved in the dynamics. The development of new and improved experimental and theoretical methodologies is being pushed in this direction. An illustrative example of the spectacular progress being made in ultrafast femtochemistry and biology is reviewed in ref. [10], which discusses the progress in time-resolved circular dichroism experiments for probing the evolution of biomolecular conformations. This combines high temporal resolution and structural sensitivity, which can now be studied down to the femtosecond timescale, providing crucial

information on the structure–function relationship. Recent progress in ellipsometry made in this context to address processes with ultrashort timescales is presented in this article.

Beyond model molecules, ultrafast experiments can address the photoinduced dynamics, where building blocks as well as advanced molecular structures can be investigated for possible applications in photovoltaics or catalysis. In ref. [11], ultrafast transient absorption and emission spectroscopies provide direct information on the photoinduced electron/energy transfer in dye-fullerene structures (BODIPY-C60-distyryl-BODIPY, BODIPY-C60, and distyryl-BODIPY-C60). While photoexcitation in dyads leads to one-electron and two-energy transfer steps from BODIPY to the fullerene, in the triad, additional energy transfer processes from BDP to DSBDP are observed. To study the multiple steps of complicated photoinduced reactions in a chemically complex molecular structure, the combination of multiple spectroscopies over an extended wavelength provides suitable probes. In ref. [12], the photophysical properties of dicyanoanthracene molecules adsorbed on the surface of zeolites have been investigated by combining picosecond transient emission, femtosecond transient absorption IR vibrational and steady-state UV–Vis spectroscopies. The authors show that the photoexcitation of the molecule leads to the formation of a localized excited state that rapidly evolves into two distinct exciplex species, identified by the frequency of the CN stretching mode. The results illustrate the possibility to develop novel photoactive materials. The extension of the spectral domain of time-resolved spectroscopies and probes also covers more extreme wavelength such as THz, as in ref. [13], where an ultrabroadband THz platform for time-resolved THz spectroscopy based on air–laser interaction is developed, providing ultrashort and intense THz pulses of 50 fs. On the other side of the frequency spectrum, the development of ultrashort pulse sources at high photon energy is also a source of new challenges, and crossing the picosecond barrier encountered in radiolysis is one of them. Here, ultrafast science might offer new means to observe the first instants following the interaction of ionizing radiation and molecules in condensed phase. In ref. [14], the computational aspects of this question are presented though the development of real-time density functional theory (DFT) calculations to simulate the electronic responses of molecular systems to ionizing radiation. Ehrenfest dynamics is used to simulate nuclear dynamics in the ground electronic state, and the authors discuss how charge transfer processes and nuclear quantum effects can be included in the simulations.

5 Ultrafast dynamics in materials and nanostructures

Controlling and monitoring the properties of materials using ultrashort light pulses is one of the expanding topics in materials science. This benefits from newly available, ultrashort pulse sources extending over a broad wavelength range, which provides a new means to observe the dynamics of all the particles and quasi-particles and their couplings, as well as the global transformation of materials. This field is now mature enough to investigate the dynamics of materials in a highly controlled environment and to approach the exotic behavior of complex materials. In ref. [15], the authors address time-resolved photoinduced ultrafast phase transition under a controlled thermodynamical environment, showing how pressure affects the out-of-equilibrium photoresponse of V_2O_3. The study of quantum materials on the ultrafast time scale is also an exciting field requiring elaborate probes, such as in ref. [16], where polarization-dependent time- and angle-resolved photoemission spectroscopy (ARPES) is used to analyze the orbital character of bands near the chemical potential for the Dirac semimetal $BaNiS_2$ for different crystal orientations. Other properties such as magnetism are also being addressed on the ultrafast time scale. In ref. [17], time-resolved X-ray magnetic circular dichroism spectroscopy is used to study the laser-induced quenching of Gd $4f$ magnetic order in different Co_xGd_y alloys, revealing that the time scales vary strongly, with a multiple-step process in which the magnetic moment loss occurs within the first picosecond before electron equilibrium, followed by a slower rate that depends on the alloy composition. In ref. [18], time-domain Brillouin scattering is used to study the possibility of inducing a coherent phonon wavepacket in a glass substrate by exciting a deposited layer of silver nanoparticles. Generation and propagation over distances of several micrometers are observed, as well as attenuation owing to the interaction with the thermal phonon bath. Progress is also being made on the theoretical side, as in ref. [19], where a first-principles formalism is used to calculate the second- and third-order susceptibilities for the electrooptic effect, including the ionic contribution, and electric-field-induced second-harmonic generation, for which TDDFT is used to calculate this nonlinear optical process. In ref. [20], the femtosecond-laser-induced evolution of the α-quartz bandgap was calculated using time-dependent density functional theory, revealing an ultrafast decrease of the bandgap during a 15-fs laser pulse. Ultrafast interactions in solids have become a common tool for material structuration, and emerging projects deal with the manipulation of spatial properties of light to improve the structuration process. In ref. [21], the authors analyze the interaction between an ultrafast Bessel beam propagating inside dielectrics creating a nanometric plasma, using particle-in-cell simulations. They describe the dynamics of the electrons heated and accelerated up to keV energies. In ref. [22], the authors demonstrate that curved line laser foci can be used to combine cutting and edge shaping of glass in a single process.

6 Secondary sources: electrons, protons, and photons

When the electromagnetic field becomes dominant over the coulombic interaction in atoms, the laser is able to drive the particles' wavefunction. This lies at the core of the development of secondary sources that emerge from the understanding and also the control of laser–matter interactions in the intense-field regime. Indeed, ultrashort laser pulses, with high intensity or peak power, favor nonlinear processes but also quantum effects such as tunnel ionization, and relativistic effects in the electron acceleration process. Adequate combination of all these effects leads to the production of ultrashort electron, proton, or photon beams. During recent decades, commercial lasers have become increasingly reliable, providing higher repetition rates, higher energies per pulse, and a greater degree of control. In ref. [23], laser-wakefield acceleration is studied in the single-cycle pulse regime, where the actual waveform of the laser electric field matters. The authors observed a carrier envelope phase controlled variation of the electron beam pointing and charge, showing the possibilities offered by controlling the exact light electric field in the production of ultrashort electron beams by laser–plasma acceleration. In ref. [24], the authors use phase-space methods to model and simulate spin-polarized plasmas. They study stimulated Raman scattering in a circularly polarized electromagnetic wave interacting with a dense electron plasma. The production of THz radiation from the intense ultrafast interactions is also a growing field of interest. In ref. [25], the generation of THz radiation produced by the propagation in air of a two-color filament using various polarizations is studied, revealing the role of the nonlinearity associated with stimulated Raman scattering. In ref. [26], two-color laser-driven generation of intense terahertz (THz) pulses in the presence of an external strong magnetic field is presented, showing the importance of the polarization and configuration to generate intense radiation. Finally, even more extreme laser intensity offer a new perspective in the study of exotic quantum processes. In ref. [27], the author discusses how petawatt laser-based experiments could be used to reach intensity regimes beyond which strong-field quantum electrodynamics could be studied. To break the current limitations, the authors propose to use a "curved relativistic mirror" based on a relativistic plasma mirror and demonstrate that it potentially gives access to extreme light intensities relevant for QED.

7 Conclusions

Recent developments are pushing experiments at extreme wavelengths (X-ray, XUV, and THz) to improve time resolution (eventually reaching the attosecond regime) and spatial resolution (element sensitivity). This includes the development of light or particle ultrafast sources with topologically controlled properties including complex polarizations and phase distributions. In all phases of matter, the use of a broad range of wavelengths would allow for the probing of all particles and quasiparticles involved in out-of-equilibrium processes. In parallel, new photocontrol strategies are emerging, offering the possibility to investigate phase transitions, quantum materials, as well as small isolated quantum systems. This requires the development of new experiments with more sophisticated spectroscopies, or with a controlled environment. Dealing with increasingly sophisticated experiments is also accompanied by improved theories that can tackle the challenge of dealing with the dynamics of all particles, theirs couplings, and the interaction with radiation. In parallel to these fundamental aspects, laser-based technologies such as ultrafast laser machining, is revealing new challenges. New frontiers in quantum physics can be reached with ultrafast lasers when ultrahigh time resolution and ultrahigh intensity can be reached. This special issue provides an overview of the premises of these developments.

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