Light by design: emerging frontiers in ultrafast photon sciences and light–matter interactions

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Abstract

Photon sciences and technologies establish the building blocks for myriad scientific and engineering frontiers in life and energy sciences. Because of their overarching functionality, the developmental roadmap and opportunities underpinned by photonics are often semiotically mediated by the delineation of subject areas of application. In this perspective article, we map current and emerging linkages between three intersecting areas of research stewarded by advanced photonics technologies, namely light by design, outlined as (a) quantum and structured photonics, (b) light–matter interactions in accelerators and accelerator-based light sources, and (c) ultrafast sciences and quantum molecular dynamics. In each section, we will concentrate on state-of-the-art achievements and present prospective applications in life sciences, biochemistry, quantum optics and information sciences, and environmental and chemical engineering, all founded on a broad range of photon sources and methodologies. We hope that this interconnected mapping of challenges and opportunities seeds new concepts, theory, and experiments in the advancement of ultrafast photon sciences and light–matter interactions. Through this mapping, we hope to inspire a critically interdisciplinary approach to the science and applications of light by design.

1. Foreword

The search for vanguard photon- and charged particle-based instrumentation has extremely high societal implications because it underpins myriad groundbreaking scientific, technological, and medical advancements [1, 2]. Ultrafast photon sciences and light–matter interactions lie at the heart of emerging scientific and engineering frontiers in life [3–8], information [9–11], universe [12, 13], and energy [14–17] sciences by ushering in unique tools, methodologies, and techniques that hold up the possibility of studying physical phenomena occurring at elementarily fast timescales. This is a foundational strategy to study physical systems because the ab initio photo-reactivity and electronic, atomic, and molecular response influence or dictate functional properties of mesoscopic systems.

Over the last two decades, groundbreaking discoveries have been enabled by the development of advanced photon- and charged-particle sources, such as few-cycle optical lasers [18, 19], high-power terahertz sources [20–22], high harmonic generation [23–25], ultrafast electron beam sources [26, 27], and x-ray free electron lasers (XFELs) [28–30], among others. These instrumentation and technological advancements have also stewarded novel methodologies to study ultrafast phenomena, including advanced extreme ultraviolet (XUV) spectroscopies [31, 32], ultrafast electron diffraction (UED) [26, 33], and femtosecond x-ray diffractive imaging and crystallography [34, 35]. Because of their foundational functionality, the scientific and developmental roadmap of next-generation photon and charged-particle instrumentation is bound to be formulated around the global challenges and opportunities of our times.

Recording stop-action events involving attosecond electronic transitions and beyond is the hallmark of today’s frontiers in ultrafast phenomena and quantum mechanics. The importance of studying the succession of these dynamics—i.e. electronic (de)excitation in attosecond timeframe, atomic and molecular in the
femtosecond range, physical chemistry in the picosecond, and so on—lies in that they may provide insight into (and eventually determine) how complex systems will function [36, 37]. Ultrashort photon wavepackets, from terahertz to x-ray, and charged-particle beams can be used to glimpse into ultrafast transitions in soft and hard condensed matter, chemistry, and biology. For instance, measuring and controlling the coherence and dephasing of electronic core and valence band excitation, or rovibrational motions of molecules and solid-state materials is integral to chemical dynamics [14, 15], which influences biological functions, material properties, and provides a means to control chemical reactions. In recent years, XFELs and UED have materialized remarkable breakthroughs in chemistry, life, and energy sciences. To name one particularly impressive area of application, we now have a much more complete understanding of the water oxidation cycle in photosynthetic reaction centers [5–8], a fundamental process enabling life on Earth and opening new technological avenues to harvest energy. Another remarkable example is the observation of light-triggered molecular response, such as electronic and nuclear rearrangements [14, 38, 39], which provides a means to understand processes relevant to materials science, chemical engineering, and biotechnology.

In the realm of quantum optics, structured photonics lay the foundation for the control and delivery of information using photons by exploiting their coherence and various degrees of freedom: amplitude, linear momentum, and spin and orbital angular momenta. In the last few decades, artificial structuring of light has revolutionized now well-established technologies such as optical communications [40, 41] and optical trapping [42, 43]. More recently, quantum information sciences (QISs) have ramped up the interest in using photons’ abundant properties for mimicking Hamiltonians of many-qubit systems or neuromorphic computing, for example. Zhong et al recently claimed quantum supremacy using photons using 50 squeezed states, which could be prepared deterministically and detected at the single-photon level [44]. Arrazola et al have also just presented a many-photon quantum circuit design and implementation in integrated nanophotonics [45].

Advances in structured photonics with programmable orbital and spin angular momenta [46–48] will continue to feed unique light properties such as space-time varying vortices and strongly localized vector fields, which can be exploited adaptively for quantum manipulation and ultrafast switching of topological materials [49, 50]. Because many ultrafast processes in correlated systems are field-phase sensitive, high-precision phase and timing control techniques, now achieving few-attosecond jitter [51, 52], have also become instrumental to coherently control the nano- and macroscopic properties of quantum materials. These photon sources are also capable of tailoring the 6D phase–space distribution of charged particle beams [46, 53–56] and thus have been proposed and employed to augment the brightness of ultrafast electron and secondary high-energy photon emission, e.g. x-ray or gamma-ray, sources [54, 55, 57, 58].

At extreme photon energies and intensities, XFELs represent the flagship of photon science instrumentation [28, 59], which in the short time since their advent have revealed conformational dynamics in biomolecules and ultrafast chemistry [4–8, 15, 16] at atomic-level spatial and femtosecond temporal resolutions. Motivated by their broad relevance, public and private investments worldwide have rendered facilities unifying quantum and nonlinear optics and laser-matter interactions to usher in advanced particle accelerators and XFEL modalities and expand our knowledge in ultrafast atomic and molecular physics and biochemistry [6, 8, 60–62]. Since access to these instruments is paramount to the democratization of their

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**Figure 1.** Research and development roadmap of the three intersecting areas of research in ultrafast photon sciences and light–matter interactions.
benefits, much effort is also currently dedicated to the investigation of downscaled alternatives to these technologies by tapping into free-space [53, 54], terahertz [63, 64], hybrid [65, 66] and on-chip [67, 68] particle accelerators as well as plasma-based x-ray sources [69], and proven their competitiveness in scientific [70–72] applications.

In this perspective article, we will map emerging frontiers in ultrafast photon sciences and light–matter interactions (figure 1) by addressing the current state and future direction three intersecting areas of research in the science and applications of light by design: (a) quantum and structured photonics, (b) light–matter interactions in accelerators and accelerator-based light sources, and (c) ultrafast sciences and quantum molecular dynamics.

2. Area 1: quantum and structured photonics

The archetypical objective of quantum photonics is to investigate strategies and technologies capable of coherently controlling nano-, macro-, and meso-sopic properties of technologically relevant quantum materials and systems using light. In this section, we will briefly review emerging sub-areas of research that fit within this general aim.

Structured and integrated photonics can open new possibilities for multi-dimensional control of quantum materials and laser-particle interactions. A simple and perhaps helpful conceptualization of structured photonics is as spatio-temporal light bullets with distinct intensity, polarization distribution, or wavefront, where the combination of these properties can be arranged to render complex and adaptable spin and orbital angular momenta beyond classical optics. Over the past decades, artificial structuring of light has undergone a remarkable evolution to produce advanced angular momenta and vector beams exhibiting unique properties such as optical vortices or 2D topological charge. Unconventional ways of thinking about light structure are inspiring new families of electromagnetic fields that circumvent previously unquestioned behavioral properties. More specifically, dynamically adaptable spatio-temporally variant topological states of light have the potential to enable QIS technologies based on photon distribution systems and selective light–matter interactions, including Floquet insulators [73], photonic skyrmions [74] or 2D metasurface polaritons [75]. This is why future structured photonic architectures could prove to be transformational in atomic and molecular physics, quantum electrodynamics (QEDs), relativistic and nonlinear optics, and particle physics.

One of the central limitations abridging their use in these and other research fields is the ability to generate light with real-time adaptable structure and, in some cases, at high intensities and peak power. Spatial light modulators are one of the most outsourced devices partially capable of attaining to this need, although they present severe average and peak power limitations in many applications [76]. An alternative power-scalable solution was recently demonstrated consisting of coherent space and frequency combs [46, 77], where all degrees of freedom defining light properties of each comb can be defined and controlled independently. In this scheme, complex states of light can be synthesized either by collapsing all combs in free space or a guided geometry, such as distributed fiber laser arrays or on-chip integrated photonics, where the complexity and finesse of light structure are ultimately restricted by the number of combs. To name one imminent prospective application, controlling electronic coherence is a long-sought-after goal in atomic and molecular, and condensed matter physics. Photo-excitation pathways can be exploited to coherently convert photons into charge excitations and electronic phase transitions for quantum and neuromorphic computing [78]. Unfortunately, coherence is often lost in extremely fast timescales due to incoherent fluctuations. Poised with this challenge, structured light technologies, and in particular, those exhibiting adaptable topographic charge together with advanced ultrafast x-ray and electron sources could be exploited to study and control decoherence mechanisms of various correlated materials such as twisted graphene, 3D direct semimetals, and 2D topological materials.

Measuring and detecting quantum phenomena, such as photon–photon correlation systems, is another major challenge in quantum optics. Especially in hyperspectral dynamics involving different elementary particles or vastly dissimilar energies of same-type particles, studying photon–photon correlation has direct implications in precision metrology and cross-particle transducers. Technologies capable of detecting and cross-correlating wave-packets with single-digit attosecond or single-particle level accuracy could prove to be game-changers in high energy physics and ultrafast sciences, especially if these technologies can operate at high data acquisitions rates beyond the gigahertz. In the realm of QIS, one could study how photon–photon interactions—for example, from an x-ray or terahertz photon to an optical photon—are mediated by novel materials that encode single-photon quantum states of light as a means to mimic quantum computation schemes, such as logic and arithmetic operations.

A plausible implementation of such a technology could rely on balanced optical cross-correlation [79], which has emerged as the current gold standard in timing control with the best demonstrated residual timing
jitter of ten as [80], ultimately only limited by quantum noise. The technique could be instrumental to understand memory effects in biological and material sciences, and ultimately control (de)coherence mechanisms in correlated systems.

Another key ingredient of a prospective implementation could rely on high-precision optical frequency combs. The precision of frequency measurements has improved by orders of magnitude [81] since their invention was awarded the 2005 Nobel Prize in physics. The most recent demonstration at telecom wavelengths shows a record 2.6 as residual timing jitter [51] of optical combs using a feed-forward carrier-envelope phase stabilization heterodyning technique [82]. But further progress in achievable optical precision requires careful investigation of both quantum noise and coherence degradation of the laser comb, which arises due to nonlinear optical processes and formulating pertinent mitigation strategies. As critical elements of the clockwork for reading out optical clocks, quantum-limited frequency combs could also offer dramatic improvements on precision standards, thereby constituting a potential leap in several fundamental tests in physics concerning drift of fundamental constants, general relativity, and QED, as well as paving the way for a replacement of the current microwave frequency standard. Last but not least, quantum-limited frequency combs in conjunction with integrated structured photonic architectures could also enable novel implementations of advanced spectroscopic techniques such as multi-dimensional dual-comb spectroscopy, anisotropic molecular fingerprinting, and coherent LIDAR technologies, among others.

3. Area 2: light–matter interactions in accelerators and accelerator-based light sources

Unifying laser and accelerator physics has been essential for the development of future accelerators, light sources, and other scientific instruments due to increasingly synergistic advances at the cross-section between these two fields. This has materialized advanced facilities around the world, such as XFELs for ultrafast photon sciences, and electron and ion sources for high energy physics and fusion energy sciences, among many others. In addition to long-standing uses in photoinjectors, recent applications to XFEL seeding, laser-based electron beam shaping, on-chip accelerators, and more have highlighted the need for research into laser-particle interactions. In the pursuit of undertaking this ever-increasing need, and to open new opportunities in next-generation accelerator and light source technology, we review some of the most recent laser-particle interaction concepts and techniques in this area.

Advanced laser-particle interactions are at the heart of the next generation of scientific instrumentation for ultrafast sciences because of an untapped potential to generate and control charged particles and render, for example, ultrahigh spectral brightness electron and x-ray pulses [83] or attosecond hard x-ray frequency combs. The first factor limiting electron beam brightness in accelerators and accelerator-based light sources, including XFELs, is the brightness of the electron source itself [84], namely the photoinjector. There are two general strategies to increase the photoinjector brightness: increasing the accelerating gradient to increase the extractable charge density and reducing the transverse electron energy spread [85, 86]. The most salient quantity to characterize the transverse energy spread is the intrinsic or thermal emittance of the photocathode. This emittance imposes a lower limit for the normalized emittance that can be generated by a photoinjector, and as a consequence, the upper limit for the brightness in a linear accelerator and accelerator-based light source. That is to say, there are multiple challenges and opportunities tethered to achieving bright electrons already from the photoemission process itself and in the few millimeters thereafter.

First, ultrafast and high-power laser physics play a key role in the generation of bright electron beams via photoemission. For instance, the 6D phase–space of an electron beam is significantly influenced by the spatial, spectral, and temporal electromagnetic laser pulse distribution. Therefore, the role of laser shaping in controlling photoemission for e-beam brightness and phase–space control is paramount to all photoinjectors, especially those operating at high current and high average powers, which makes high-fidelity high-power shaping central to the production of high brightness beams beyond XFELs, including compact accelerators [87] and particle colliders [88]. Notwithstanding the foundational role of photoinjector laser systems, the production and high-fidelity characterization of temporally tailored photoexcitation laser pulses, typically in the ultraviolet (UV) range, is challenging under traditional photoinjector configurations due to reduced available UV bandwidth and material damage [89].

Recent theoretical studies and experimental results in nonlinear upconversion techniques—including sum-frequency generation [58] and four-wave mixing [90]—and new photoemission regimes [91], promise to consolidate next-generation photoinjector technologies to overhaul long-standing challenges in the generation of bright electron beam sources, which include in addition to shaping, efficiency, stability, and high-power handling. Some of these prospective solutions enable arbitrary shaping of the upconverted pulses in real-time [92], a long-sought-after challenge to control and improve the electron beam characteristics in photocathode emission. Such a capability would also enable machine-learning-based application in electron phase–space control for fast and fine optimization and multiplexing at above-kilohertz repetition rates.
At the charged-particle control and acceleration level, light bullets by design can impose high-precision microcorrelations onto charged-particle beams. For example, periodic electron modulations have been postulated to yield fully coherent and mode-locked hard x-ray pulses [93], i.e. few-attosecond duration x-ray pulse train, which would epitomize the same radical advancement in ultrafast sciences the mode-locked optical laser has brought about since the 1980s. Among some recent developments, relativistic vector, orbital angular momentum, and other non-conventional laser beams have demonstrated some advantages in the conditioning or tailoring of electron beams [19, 55, 94]. To take one of these examples, a first-order orbital angular momentum beam was recently demonstrated to reduce the energy spread of electron bunches, which are very susceptible to instabilities, and avoid degrading the coherence, spectra, and performance of XFEIs [94]. Compact accelerators can also benefit from light bullets by design and rendering distributed photon sources in the form of structured photonics. The generation of relativistic vector beams had demonstrated free-space acceleration [53], which circumvents the use of any accelerating structure or guiding medium, and thus promises to produce high-quality Heisenberg-limited relativistic electron beams [54]. Alternatively, dielectric-based on-chip accelerators have also ushered in unprecedented accessibility and cost promise [67, 68] by miniaturizing traditional radiofrequency accelerators down to optical wavelength dimensions. However, staging of linear accelerators and efficient electron–photon coupling stand out as some of the primary challenges in the way before their widespread applicability and market release. Bringing laser-driven electron sources outside of the facility complex to an university-, industry- or hospital-level operational capacity could efficiently render few-attosecond megaelectron volt-level electron wave-packets suited for attosecond ultrafast diffraction efficiency, QIS, and medicine, such as targeted microdose radiation therapy, among myriad other opportunities.

4. Area 3: ultrafast sciences and quantum molecular dynamics

Some of the emerging technologies and methods laid out in the prior two areas bear a unique inquisitive power to excite and probe dynamic molecular processes in atomic, molecular, and condensed matter physics. These tools can be used to deepen our understanding of fundamental processes underpinning energy and life sciences. In this topical area, we examine new opportunities in two relevant molecular systems that bracing many of these processes: water and carbon dioxide.

Liquid water is the single most important medium in which highly consequential chemical and biological processes take place [95, 96]. Water is often equivocally regarded as a passive medium but it conversely shows an unique, active, and ubiquitous behavior at different temperatures and environments ranging from an isolated molecule to small clusters and up to bulk. As such, water plays a crucial role in a wide variety of chemical reactions, such as glucose isomerization [97] and photoionization of indoles among others, as well as in biological processes, such as twisting of DNA double helix [98] and the recognition of DNA sequence [99]. Recently, terahertz absorption spectroscopy of solvated biomolecules has shown that water is a key factor in enzymatic reactions and protein folding, providing microscopic visualizations of the absorption dynamics at picosecond timescales [100]. These dynamics are not yet fully understood since they are highly susceptible to the H-bond network, orientation, relaxation rates, and density of liquid water.

Most chemical processes in which water plays an active role are triggered in very fast timescales. Water librations, a fundamental molecular vibrational mode in the far-infrared (FIR) and terahertz range, are investigated as proxy mechanisms for kinetic and first-few hydration layer activation compared to traditionally employed OH-resonances, i.e. a means to effectively activate quantum mechanical processes in water and transfer radiated energy to liquid using librational molecular excitation rather than atomic vibrations. This is because librations are the hindered molecular rotations that are directly affected by the H-bond with other molecules. Hynes et al have extensively investigated the energy relaxation mechanism from librations to other internal vibrations [101, 102]. Due to the strong terahertz cross-section with librations, it is possible to excite and transfer significantly large energy to liquid water using moderate intensities in the order of 108 W cm−2 and above in the terahertz frequency range. At this and higher intensities, terahertz radiation can weaken or fully disrupt the H-bond and simultaneously allow permanent dipoles of water to oscillate with the electric field.

As a direct consequence of this prospect, femtosecond and picosecond FIR and terahertz pulses could control aqueous environments with an extraordinary dynamic range, from T-jumps in the sub-K up to supercritical-like water structures, to universalize thermally activated ultrafast structural dynamics and new ultrafast solvation chemistry [103]. Furthermore, strong librations can also produce unique phase states of water, including supercritical [104] and supercooled water, which could be relevant to study Earth and exoplanet atmospheric conditions in a laboratory. Understanding these dynamics could unlock a vast
ensemble of chemical reactions in hydration shells that are not possible to study today or to develop new atmospheric and geo-engineering technologies.

The nonlinear photo-excitation and photo-catalysis of carbon-based systems is another sought-after area of research. Photo-dissociative pathways of carbon dioxide have been extensively studied because of highly consequential functions in Earth and planetary sciences [105, 106] and physical, biological, and environmental chemistry [107–110]. Linear absorption in the vacuum ultraviolet (VUV) and XUV range can access high-lying Rydberg states in CO$_2$ [111]. In these photoexcited states, generally $\sim$11 eV above the minimum of the ground electronic state, the large internal energy opens various interesting dissociation and isomerization pathways [112–114], including roaming leading to unusual photochemical products [115].

But in the nonlinear absorption regime, the response of a molecule upon multi-photon excitation can reveal new intra-molecular reaction mechanisms occurring on attosecond timescales. The response can be very sensitive to specific energy and phase features of the photoexcitation wave packet, thus providing a mechanism to control which molecular excitation states are populated and when. That is, the nonlinear photoexcitation response of carbon dioxide, as well as other di- and tri-atomic molecules, and subsequent photofragment production can unveil the complex interplay between multi-photon excitation, excited states, and internal energies of the molecular and ionic fragments. For example, nonlinear Fourier transform spectroscopy [31] can reflect the dynamics of electronic wave packets created via the two-photon excitation in the CO$_2$/CO$_2^+$ system, which may carry information on the energy differences between the ionizing and dissociating states of molecular ion [116]. Alternatively, ultrafast x-ray or electron scattering methods can also be employed to visualize linear and nonlinear photoexcitation and dissociation dynamics of the neutral parent molecule which may render reciprocal space dynamical information of the intra- and inter-molecular electron distributions, inter-atomic distances, and bond angles of the VUV/XUV-excited Rydberg states of all atomic fragments and species.

In the near future, studying the role of photocatalytic reactions in splitting carbon dioxide via nonlinear excitation and precisely mapping relevant intermediate and final dissociation steps could also reveal strategies to steer and trace the electronic and molecular motion of CO$_2$ to increase the photo-dissociative yield to levels where decarbonization strategies and negative emission technologies based on nonlinear photocatalysis may become reality in the face of climate change.

5. Closing remarks

In this perspective article, we have presented the emerging scientific and technological frontiers of three intersecting and often semiotically distinct research areas. First, we have reviewed quantum and structured photonics, and indirectly nonlinear and ultrafast optics, as a self-standing field primarily founded on light by design with applications in attosecond metrology and precision frequency combs for applications in QIS and QED, and concerning the study of molecular dynamics relevant to applications in chemical, environmental, and biological physics. We have also examined light by design as enabling new possibilities in the development of scientific instrumentation, such as compact XFELs and control and manipulation of charged-particle beams in accelerators and light sources, which in turn become powerful tools to observe ultrafast molecular dynamics and understand the processes that underpin material, biological, and chemical properties. Last, we have presented some prospective applications in life sciences, biochemistry, and environmental and chemical engineering, founded on a broad range of photon sources, from terahertz to x-ray. We hope that this interconnected mapping of challenges and opportunities seeds new concepts, theory, and experiments in the advancement of ultrafast photon sciences and light–matter interactions.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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