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ABSTRACT

The science of x-rays is by now over 125 years old, starting with Wilhelm Röntgen's discovery of x-rays in 1895, for which Röntgen was awarded the first Nobel Prize in Physics. X-rays have fundamentally changed the world in areas, including medical imaging, security scanners, industrial inspection, materials development, and drugs spectroscopy. X-ray science has been so far responsible for over 25 Nobel Prizes in Physics, Chemistry, and Medicine/Physiology. With x-ray generation being a highly commercialized, widely adopted technology, it may appear that there is little left to discover regarding the fundamentals of x-ray science. Contrary to this notion, recent years have shown renewed interest in the research and development of innovative x-ray concepts. We highlight, in this Perspective, promising directions for future research in x-ray science that result from advances in quantum science and in nanomaterials. Specifically, we describe three key opportunities for advancing x-ray science in the near future: (1) emerging material platforms for x-ray generation, especially 2D materials and their heterostructures; (2) free-electron-driven emission of entangled photon–photon and electron–photon pairs for x-ray quantum optics; and (3) shaping free-electron wavepackets for controllable x-ray emission. These research directions could lead to improvements in x-ray resonance fluoroscopy, high-contrast x-ray imaging, stimulated coherent x rays, x-ray superradiance, and other prospects for x-ray quantum optics.

I. INTRODUCTION

X-rays are indispensable in modern society, with widespread uses in medical imaging, industrial quality inspection, security scanning, and the pursuit of fundamental research.[1,2] While x-ray tubes are ubiquitous in medical, industrial, and scientific applications, recent decades have also witnessed the rise of intense, tunable, and directional x-ray sources in the form of enormous, expensive synchrotron, and free-electron laser facilities.[3–5] By producing ultrashort x-ray pulses, these facilities open the doors to spectroscopy of material dynamics and biological processes.[6] Moreover, the coherence of such x-ray sources enables higher resolution imaging through phase contrast techniques,[7] safer medical imaging by reducing the needed dosage,[8,9] and next-generation security inspection of microchips.[10] The size and expense of synchrotrons and free-electron lasers, however, have been an obstacle to their widespread adoption in commercial and medical applications. Soft x rays have proven useful in biological imaging, especially in the water window where water is transparent to x rays, facilitating the study of organic compounds and biological specimens in their natural aqueous environment.[11] Hard x rays are especially important for medical imaging and security scanners, with even harder x rays bordering on gamma-rays being used in large-scale industrial applications.

Free electrons are central to x-ray generation for a variety of reasons. They are readily produced and accelerated to relativistic velocities at relatively low kinetic energies, enabling them to serve effectively as highly nonlinear optical media. This fact is perhaps most readily seen in inverse Compton scattering, whereby light scattering off a moving electron can be Doppler up-shifted to a much higher frequency.[12] Based on similar principles, x-ray synchrotrons and free-electron laser facilities generate x rays by accelerating electrons to ultrarealistic kinetic energies (e.g., 100's of MeV to few GeV), before sending these electrons through magnetic undulators.

The drive for ever more compact, efficient, and high-quality x-ray sources has led to substantial innovation. The prospect of shrinking the electron accelerator stage in free-electron x-ray sources (such as x-ray free-electron lasers) has motivated the study of high-gradient acceleration mechanisms, including laser-driven plasma acceleration[13–15] and dielectric laser acceleration[16–18] that use on-chip silicon photonics[19] and terahertz acceleration.[20,21] In laser-driven free-electron radiation mechanisms like inverse Compton scattering, the effective undulator period can be 100–1000 times smaller than that of conventional magnetic wigglers and undulators, allowing for x-ray emission with relatively low energy electrons, and thus more compact footprints.[22–26] In x-ray tubes, carbon nanotube emitters have been shown to possess advantages over conventional cathodes, such as improved electron beam profiling, higher beam current, and enhanced temporal stability.[31]

X-ray generation mechanisms that do not involve an external source of free electrons have also attracted much interest. The interaction of intense lasers with plasmas in high harmonic generation produces soft x-ray attosecond pulses and frequency combs.[10,27,28] Compact, high-flux hard x-ray sources have been demonstrated via laser-driven characteristic x-ray emission from solid targets.[29,30] Triboluminescence has emerged as a niche method of generating x-ray pulses on the order of 10 ns in duration.[32] It should be noted that although these techniques do not involve an external source of free electrons, they all rely on acceleration of at-least-partly free electrons by electromagnetic fields to reach relatively high kinetic energies, usually followed by a recombination event that emits x rays.

With myriad x-ray generation techniques being well-known and refined for many years, x-ray science is by now a mature research area, and it may seem that everything that is to know about it is already known. Yet, renewed interest in the field in recent years has resulted in important technical advances and proposals of innovative concepts for x-ray sources.

In this Perspective, we emphasize emerging research directions in x-ray science that open up unique opportunities for future x-ray technology and are only now accessible owing to advances in nanophotonics, 2D materials, and quantum optics. We describe how the highly nonlinear and spectrally broadband nature of the free electron allows us to leverage the versatility of nanophotonics for the manipulation and enhancement of x-ray emission. This concept relies on the already well-known abilities of nanophotonics to mold the flow of light at optical, infrared, terahertz, and microwave wavelengths. We identify three main themes of immediate interest to x-ray science: the study of emerging materials to serve as nanophotonic platforms for compact, tunable, and high-brightness x-ray generation; free-electron-driven emission of entangled photon–photon and electron–photon pairs for quantum optics; and the quantum shaping of electron wavefunctions to control the spatiotemporal profile of emitted x-rays. The overall process and prospects of free-electron x-ray nanophotonics is illustrated in Fig. 1. We also discuss prospects for future research along the way.

II. EMERGING MATERIAL PLATFORMS FOR X-RAY GENERATION

Rapid strides in nanofabrication methods have led to the discovery of an ever-widening range of materials to manipulate light on a subwavelength scale. These materials—which include plasmonic materials, 2D materials, metamaterials, metasurfaces, and topological materials (both electronic and photonic)[33–41]—have been used in the design and enhancement of classical and quantum light sources.[42–44] However, these light sources had been restricted to frequencies far below the x-ray regime, due to atomic transitions predominantly being in the visible and near-ultraviolet range, i.e., corresponding to low energy photons (∼1–10 eV). Even in cases where extreme ultraviolet (EUV) and x-ray transitions exist (e.g., core–shell transitions), the transition rates are too fast and had been impractical for extending the concepts of lasers to the EUV and x ray.

Free electrons can bridge the gap between nanophotonics and x rays as a result of one (or both) of two unique properties: the spectrally broadband nature of the charged particle's Coulomb field and the ability of a moving free electron to behave as a highly nonlinear optical medium.[45] Equivalently, nanophotonics can be understood as a proxy that converts energy from the free electrons to x-ray radiation. The optical nonlinearity of electrons has already been leveraged for x-ray generation in synchrotrons and free-electron lasers, where relativistic electron velocities Doppler-shift the centimeter-scale periodicity of the undulator into sub-nanometer x-ray wavelengths. The prospect of reducing the effective undulator periodicity to the micrometer scale, allowing for more compact setups with lower energy electrons, has motivated inverse Compton scattering designs based on infrared or optical sources.[46,47] In inverse Compton scattering, the use of Bragg structures has been proposed to guide the counter-propagating laser pulse, thereby overcoming diffraction issues in free space laser pulses and enhancing the output intensity by orders of magnitude[48] [Fig. 2(a)]. All-dielectric undulators that use dielectric structures to shape the profile of high-repetition-rate and moderate-power laser fields for electron deflection have been proposed as a means to realize table-top x-ray free-electron lasers[49] [Fig. 2(b)].

Smith–Purcell radiation, which leverages the spectrally broadband electron near-field, has also been studied as a compact source of x-ray radiation.[50] The x-ray component of an electron's Coulomb field is diffracted off a periodic nanostructure into propagating x-ray radiation.

Rapid advances in emerging materials like plasmonic platforms and van der Waals (vdW) structures have broadened the possibilities for compact x-ray free-electron sources. Two-dimensional (2D) materials like graphene can support surface plasmon polaritons with field confinements of over two orders of magnitude.[51–53] This results in effective undulator periodicities of 10–100 nm or less. The high momentum of these strongly confined graphene plasmons enables the generation of exceptionally high-energy output photons when electrons scatter off these plasmons [Fig. 2(c)]. Considering graphene plasmons of field confinement factor of 180, highly directional (<10 mrad angular spread), tunable, and monochromatic (0.25% photon energy spread) 20 keV x rays can be generated using modestly relativistic electrons of kinetic energy 3.7 MeV, in a micrometer-scale footprint.[54] This compact x-ray generation scheme bypasses the need for lengthy electron acceleration stages or extreme laser intensities. Such a device would also feature exceptional tunability—spanning the electromagnetic spectrum from infrared to hard x-ray frequencies—that is controllable in three ways: by varying the electron energy, the frequency of the surface plasmon, and the graphene doping (for example, by electrical gating).[54]

Even greater versatility can be achieved in the polariton-based free-electron x-ray source by using plasmonic metasurfaces. In particular, it has been shown that the output x-ray spectral profile can be arbitrary shaped by controlling the metasurface geometry, the electron energy, and the incidence angle of the ultrashort laser pulse responsible for inducing polaritons[55] [Fig. 2(d)]. A variant of the above

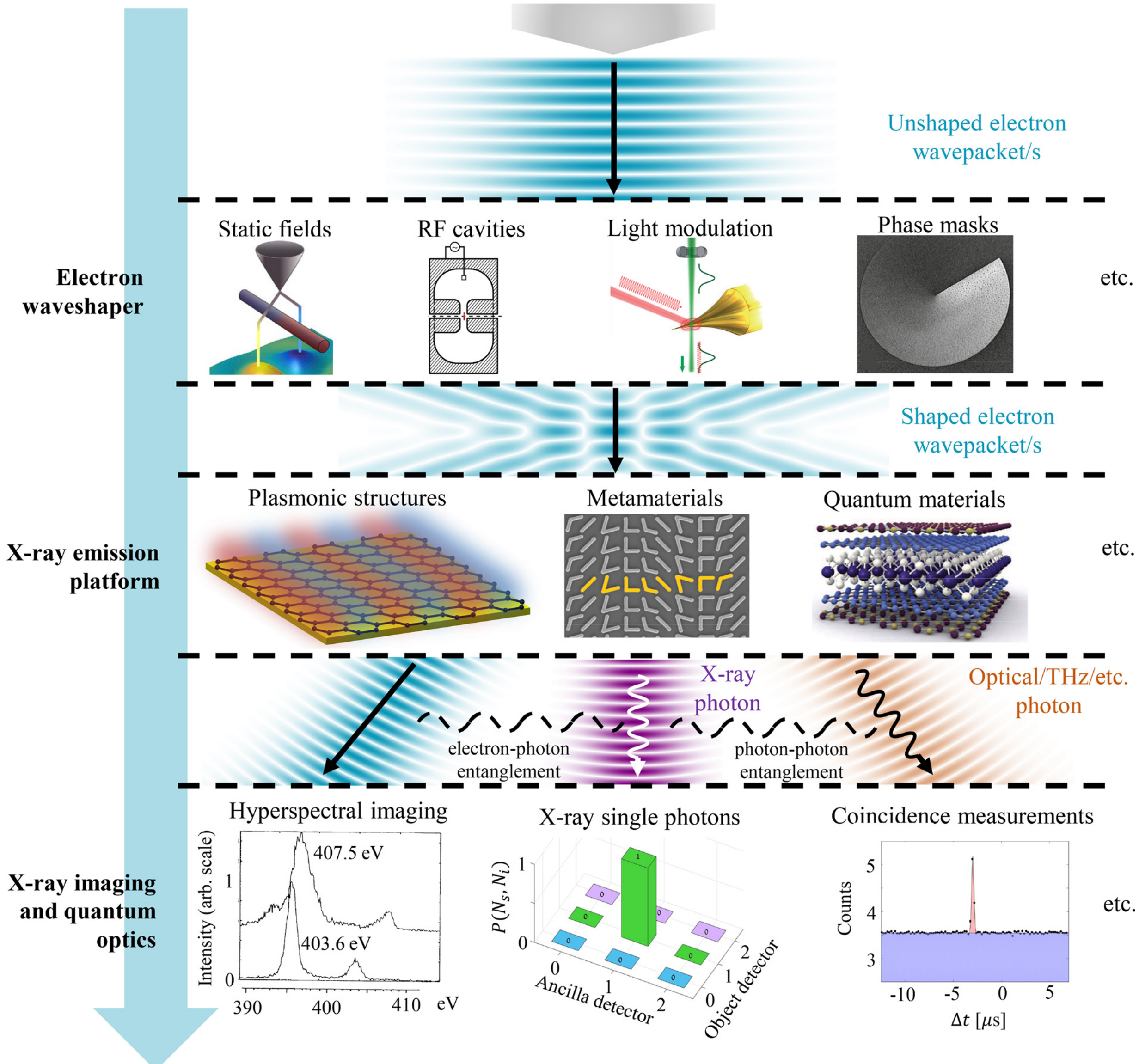

FIG. 1. Overview of free-electron-driven x-ray nanophotonics and quantum optics: shaped electron wavepackets of nonrelativistic to moderately relativistic energies interact with nanomaterials to generate x rays. The output x rays are entangled with other output particles, including the outgoing electrons and other photons or quasi-photons. Electron waveshaping and nanomaterial tunability can be used to control and enhance the x-ray output characteristics, while the entanglement can be potentially leveraged in x-ray quantum optics and imaging applications. Hyperspectral imaging: the diagram shows nitrogen K-emission spectra recorded at different x-ray photon energies (labeled), demonstrating site-selective probing by the excitation of different resonances using different input x-ray energies from a synchrotron.[157] X-ray single photons: diagram shows the probability distribution of the number of photons detected by ancilla (signal) and object (idler) detectors by using coincidence measurements for an x-ray photonic downconversion quantum source.[81] Coincidence measurements: diagram shows time difference between x-ray and electron energy loss (EEL) events in a spectroscopic coincidence experiment in a transmission electron microscope. The results show a clear temporal correlation (red peak), with uncorrelated events present as background (blue).[115] Illustration for "static fields" is used reprinted with permission from Guzzinati *et al.*, Nat. Commun. 8, 14999 (2017).[154] Copyright 2017 Authors, licensed under a Creative Commons Attribution (CC BY) license; illustration for "RF cavities" is reprinted with permission from van Oudheusden *et al.*, Phys. Rev. Lett. **105**, 264801 (2010).[120] Copyright 2010 American Physical Society; illustration for "light modulation" is reprinted with permission from Feist *et al.*, Nature **521**, 200–203 (2015).[122] Copyright 2015 Springer Nature; illustration for "phase masks" is reprinted with permission from Shiloh *et al.*, Ultramicroscopy **144**, 26–31 (2014).[136] Copyright 2014 Elsevier; illustration for "metamaterials" is used reprinted with permission from Yu *et al.*, Science **334**, 333–337 (2011).[36] Copyright 2011 AAAS; illustration for "quantum materials" is used reprinted with permission from Novoselov *et al.*, Science **353**, aac9439 (2016).[34] Copyright 2016 AAAS; illustration for "hyperspectral imaging" is reprinted with permission from J. Nordgren and N. Wassdahl, J. Electron Spectrosc. Relat. Phenom. **72**, 273–280 (1995).[157] Copyright 1995 Elsevier; illustration for "x-ray single photons" is used reprinted with permission from Sofer *et al.*, Phys. Rev. X **9**, 031033 (2019).[81] Copyright 2019 Authors, licensed under a Creative Commons Attribution (CC BY) license; illustration for "coincidence measurements" is reproduced with permission from Jannis *et al.*, Appl. Phys. Lett. **114**, 143101 (2019).[115] Copyright 2019 AIP Publishing.

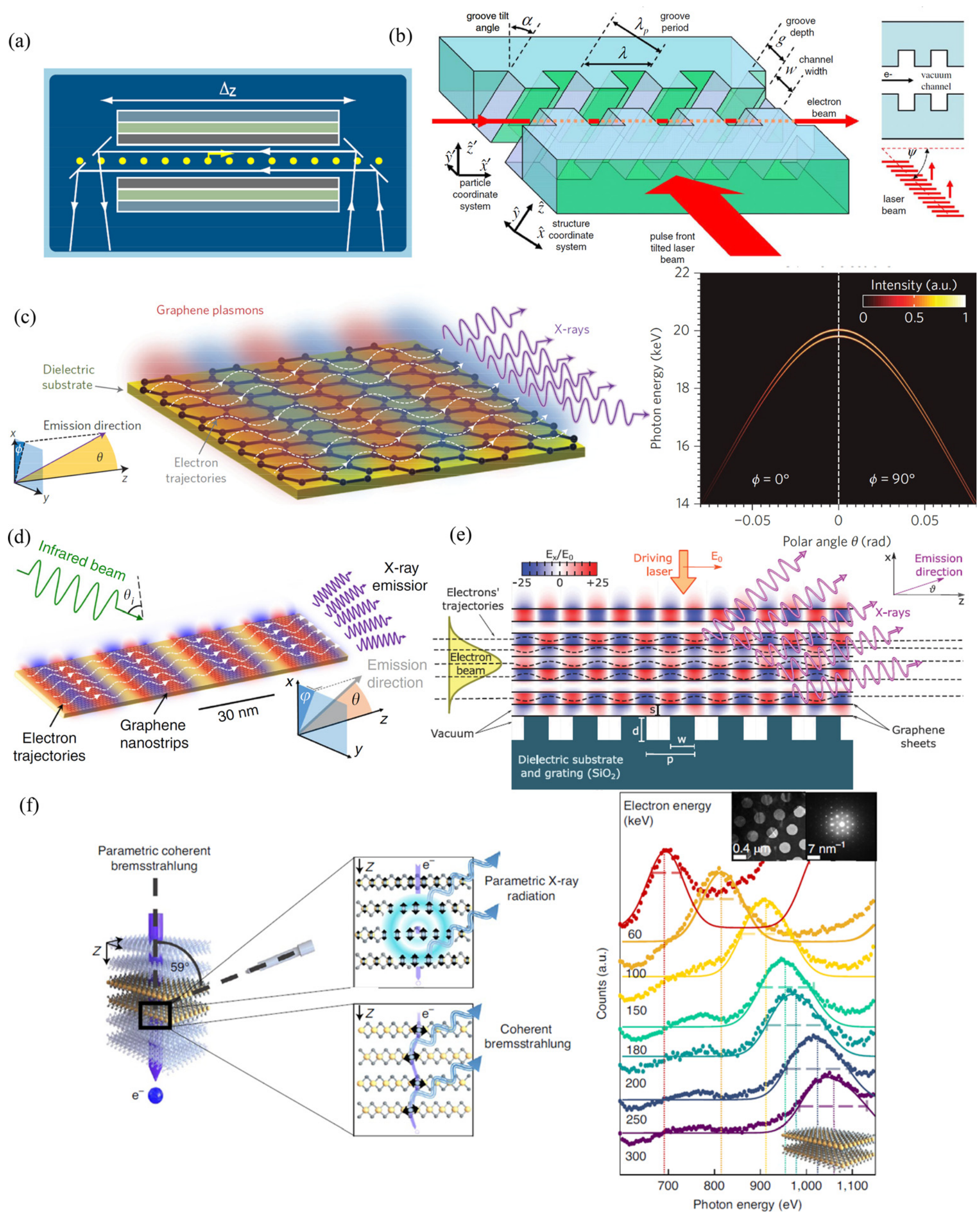

FIG. 2. Nanophotonic x-ray emission platforms. (a) Inverse Compton scattering in a Bragg waveguide, which increases the interaction length and leads to x-ray intensity enhancements of over two orders of magnitude compared to a free space laser beam; (b) dielectric laser undulator in which an incident laser beam wiggles the input electrons, causing them to emit x rays; (c) graphene plasmon-based x-ray source (left), which leverages the high confinement factor of graphene plasmons to generate tunable, highly directional hard x rays from moderately relativistic electrons (the right diagram shows the output x-ray spectral intensity for 3.7 MeV electrons interacting with graphene plasmons of free space wavelength 1.5 μ m and confinement factor 180, over an interaction length of 1.5 μ m); (d) plasmonic metasurfaces (graphene nanoribbons in this specific example) as a versatile source of multi-harmonic x rays, whose spectral content can be controlled by tailoring the near-field through the metasurface design; (e) plasmonic metamaterials as a means of increasing the interaction volume of electron–plasmon interaction, allowing x-ray intensity enhancements of over 100 times; (f) van der Waals materials as a platform for tunable, high-brightness soft x-ray generation (left), demonstrating the concept of x-ray tunability via material design on the atomic scale. The right diagram shows the output x-ray spectral intensity for different incident electron energies (labeled) on WSe_2, with insets showing the sample image (top right first), diffraction pattern (top right second), and a 3D model for WSe_2 (bottom right). (a) Figure reprinted with permission from Karagodsky *et al.*, Phys. Rev. Lett. **104**, 024801 (2010).[48] Copyright 2010 American Physical Society; (b) used reprinted with permission from T. Plettner and R. L. Byer, Phys. Rev. Spec. Top.-Accel. Beams **11**, 030704 (2008).[49] Copyright 2008 Authors, licensed under a Creative Commons Attribution (CC BY) license; (c) reprinted with permission from Wong *et al.*, Nat. Photonics **10**, 46–52 (2016).[54] Copyright 2016 Springer Nature; (d) used reprinted with permission from Rosolen *et al.*, Light **7**, 64 (2018).[55] Copyright 2018 Authors, licensed under a Creative Commons Attribution (CC BY) license; (e) used reprinted with permission from Pizzi *et al.*, Adv. Sci. **7**, 1901609 (2020).[57] Copyright 2020 Authors, licensed under a Creative Commons Attribution (CC BY) license; (f) reprinted with permission from Shentcis *et al.*, Nat. Photonics **14**, 686–692 (2020).[62] Copyright 2020 Springer Nature.

polariton-based x-ray generation schemes, using terahertz-driven surface plasmons, has been proposed.[56]

The use of plasmonic metamaterials, such as graphene metamaterials [Fig. 2(e)], provides a means of scaling up the x-ray output intensity of polariton-based undulators by over two orders of magnitude, by supporting a larger interaction volume with the electron beam.[57] The graphene-metamaterial-based x-ray source also introduces a unique paradigm for free-electron light sources where the electron mean free path is longer than the device length, relaxing the conventional requirement that electrons must travel in vacuum to avoid beam degradation through scattering events.[57]

X rays can also be generated in nanocrystals, such as carbon nanotubes, through free-electron-based processes like channeling radiation, coherent bremsstrahlung (CB), parametric x-ray radiation (PXR), and nanotube undulator radiation.[58] Nanocrystals like carbon nanotube ropes have advantages as x-ray sources through their relatively low x-ray absorption—allowing for more intense x-ray output—and more stable channeling, as compared to ordinary crystals.[58]

The concept of tunable x-ray generation by controlling material response takes on an even more exciting aspect when applied to vdW materials, whose isolated atomic planes can be reassembled into designer heterostructures made layer by layer in a precisely chosen sequence[59] [Fig. 2(f)]. Emerging types of polaritons and innovative methods of controlling them in vdW structures[60,61] imply additional ways to design versatile, compact free-electron polariton-based x-ray sources. The versatility of vdW structure design also extends to x-ray generation processes based directly on the crystal lattice itself, such as parametric x-ray radiation and coherent bremsstrahlung. Recent experiments[62] have demonstrated the ability of van der Waals materials to serve as a platform for tunable x-ray generation when irradiated by moderately relativistic electrons from a transmission electron microscope. The radiation spectrum can be precisely controlled by tuning the acceleration voltage of the incident electrons as well as adjusting the lattice structure of the van der Waals material. This experiment, thus, demonstrated the concept of material design at the atomic level, where the wealth of possibilities in designing vdW materials and other atomic superlattices opens up further areas of exploration in x-ray physics and technologies.[62] The appeal of using vdW materials for x-ray generation is enhanced by the fact that many vdW materials possess high in-plane thermal conductivities,[63] and some have higher melting temperatures compared to conventional materials. Moreover, radiation damage can be further reduced by using heterostructures combining different kinds of vdW materials.[64,65] It has also been shown that the PXR mechanism in vdW materials can be particularly designed to generate ultrashort pulses and delta-pulse trains with a controllable period, angular distribution, and polarizability.[66]

With many of these x-ray radiation stages occupying a chip-scale footprint, the application of micro-electro-mechanical-system (MEMS) is an intriguing prospect for creating highly versatile, compact x-ray sources for fluoroscopy and imaging. MEMS has already been used to realize dynamic x-ray optics that can manipulate hard x-ray pulses on time scales down to 300 ps,[67] within an order-of-magnitude of the x-ray pulse duration from many standard synchrotron sources.

The burgeoning potential of x-ray free-electron nanophotonics continues to grow with the emerging materials and methods of controlling electromagnetic fields in materials. The ability to realize highly confined magnetic fields at the surface of nanopatterned ferromagnets,[68] for instance, has motivated the investigation of ferromagnetic nanograting x-ray undulators.[69] Further possibilities remain to be discovered, for instance, in the magnetic and topological properties of natural vdW crystals[70] and their heterostructures, and in the exotic physics of one-dimensional (1D) vdW heterostructures.[71] The free electron is unique as a robust and powerful means of bridging the gap between control of physics at DC-to-optical wavelengths and control of physics at extreme frequencies like the x-ray regime.

III. FREE-ELECTRON-DRIVEN EMISSION OF ENTANGLED PHOTON–PHOTON AND ELECTRON–PHOTON PAIRS FOR X-RAY QUANTUM OPTICS

Quantum optics has been applied to improve the quality of measurements over what is possible with classical illumination, through methods like quantum imaging and quantum metrology.[72–75] However, these methods have largely been limited to parts of the electromagnetic spectrum far below the x-ray regime.

Extending quantum optics into the x-ray range has been argued to be beneficial from both a fundamental and practical point of view. From the viewpoint of fundamental quantum optics, detectors in the x-ray range are capable of resolving the number of detected photons with nominally zero background noise and a broadband unity quantum efficiency. Such capabilities are beyond the reach of quantum optics in the optical range and could, thus, enable basic tests of fundamental concepts in quantum information such as Bell-type inequalities,[76] ideas like ghost imaging,[77] and the creation of multi-photon Fock states using efficient post-selection.[78,79] Quantum imaging with incoherently scattered x rays has been demonstrated by leveraging higher-order degrees of coherence.[80]

From the viewpoint of x-ray science and applications, the introduction of quantum-optical concepts can help access undiscovered atomic-scale phenomena using the extremely short photon x-ray wavelengths.[81] X rays have been shown to be an enabling tool for nuclear quantum optics as well as studies of collective and virtual effects in the interaction of identical atoms with single photons.[82,83] For instance, it has been shown that the dynamics of Mössbauer nuclei can be coherently controlled using an x-ray double-pulse,[84] and that one-photon superradiance and Dicke state superradiance can be achieved using an ensemble of nuclei.[85–88] An ensemble of nuclei can also be used to shape the waveform of gamma-ray photons coherently, providing a source of single-photon, ultrashort gamma ray pulses with controllable waveforms.[89]

In the field of x-ray quantum optics, correlated x-ray photons were recently demonstrated using x-ray downconversion.[77,81,90–94] The mechanism has been used as a source for ghost imaging[77] and shown to generate heralded photons with sub-Poissonian statistics.[81] Instead of two entangled x-ray photons, spontaneous parametric downconversion can also be used to produce an entangled photon pair consisting of an x-ray photon and an optical photon.[95–99]

Such x-ray sources have substantial improvements in visibility and signal-to-noise ratio in coincidence detection schemes involving the two entangled x-ray photons.[81] Efficient interaction of heralded x-ray photons with beam splitters was recently demonstrated, opening the doors to innovative setups for x-ray quantum optics that involve beam splitters and single photon interactions.[100] Nevertheless, all these studies used another photon for the post-selection, which meant that the efficiency of the coincidence events was limited by the efficiency of the nonlinear process, and the efficiency of the detection of the other photon. Correlating the x-ray photon and the electron could lead to a more efficient process and benefit from the higher efficiency of electron detection.

It is noteworthy that quantum vacuum fluctuations can be used to produce x rays from free electrons through Casimir-type forces that undulate the electrons. For example, polariton-based free-electron x-ray generation is possible even in the absence of externally induced polaritons. Instead, the electromagnetic vacuum fluctuations (e.g., of polariton modes) that exist in the nanoscale vicinity of materials[101] can interact with the electron and lead to x-ray generation [Fig. 3(a)]. The interaction of these fluctuations with free electrons is equivalent to a quantum optical two-photon process in which a free electron spontaneously emits an entangled pair of a low-energy polariton and a high-energy photon. The radiated power is comparable with that from an equal-energy electron in an external magnetic field of strength on the order of 1 T. The strength of this x-ray generation process is related to the strong Casimir–Polder forces that atoms experience in the nanometer vicinity of materials, with the essential difference being that the fluctuating force here acts on free electrons, instead of neutral, polarizable atoms. The resulting x rays can be potentially shaped by controlling the nanophotonic geometry or the underlying material electromagnetic response at optical or infrared frequencies.[101] This means that concepts from nanophotonics that are used to shape the local photonic density of states in the optical spectrum (using metasurfaces, etc.) can affect the physics in the x-ray spectrum as well. For example, photonic crystals can be used to manipulate the local density of photonic states in the optical range, to indirectly control the more-elusive local density of photonic states in the x-ray range.[102]

Free-electron-driven methods of generating x rays have an inherent advantage for applications in quantum optics because they involve more than one output particle: the emitted x-ray photon is emitted in a joint entangled state with the emitted electron. The output particles are entangled to one another due to energy-momentum conservation laws, restricting the possible energy and direction of one particle when those of the other particle are known. Free-electron x-ray generation is, thus, a potential means of realizing detection schemes based on electron–photon entanglement at x-ray wavelengths. For example, it would be possible to realize heralded single-photon x-ray sources based on the electron measurement [Fig. 3(b)].

Let us discuss what are the most promising experimental platforms for exploring x-ray quantum optics. All existing studies of x-ray quantum optics have involved either x-ray tubes, which are neither tunable nor directional, or large-scale facilities like synchrotrons, which are less accessible than lab-scale setups. A free-electron-based nanophotonic x-ray source (Sec. II, Fig. 2)—such as the vdW-based x-ray source[62] [Fig. 2(e)]—could be highly complementary to both technologies, as it is not only lab-scale but also tunable and directional. It is advantageous that the brightness achieved by the vdW-based x-ray source can potentially surpass that from x-ray tubes.[62] The use of pulsed electrons (and ultrafast plasmon pulses in the case of plasmon-driven free-electron x-ray sources) opens up the possibility of sub-picosecond x-ray pulses from these nanophotonic schemes. The high-brightness, dynamically tunable and highly directional nature of free-electron-based nanophotonic x-ray sources also make them promising for generating x-ray single-photons that are heralded by measuring the electron energy loss.

The transmission electron microscope (TEM) is an excellent platform to explore quantum optical processes with free electrons. This is because it is designed to generate electron waves of high spatial coherence and sometimes also temporal coherence. Furthermore, it can usually be equipped with multiple detectors for coincidence detection of output electrons and photons. TEMs have been mainstays of scientific progress since their invention exists almost a hundred years ago. An electron incident on a material leads to inelastic excitation processes that remove energy from the electron and excite a photonic quasiparticle or another collective mode in the material (e.g., phonons, plasmons, excitons, etc.).[103] Some of these excitations later emit a photon through recombination (e.g., inner-shell excitation and de-excitation emit x-ray photons, while outer-shell electron–hole recombination emits optical photons). This process is generally known as cathodoluminescence,[104] with the cases of x-ray emission more commonly studied as part of energy dispersive x-ray spectroscopy (EDS).[105] The electron energy loss and photon emission can be quantitatively measured in order to characterize a material, through techniques like electron energy loss spectroscopy (EELS), detecting the energy loss of accelerated electrons,[106–108] and EDS, detecting x rays.[109–111]. In modern TEM instruments, both EELS and EDS can be used to analyze the sample simultaneously.[112,113] Previous works,[114] including important recent advances,[115] show that exploiting the intrinsic coupling between the EELS and EDS signals in coincidence measurements can provide spectroscopic information with a significantly suppressed background. Such coincidence measurements are the first to use the entanglement between the free electron and the excitation it creates, which has many more promising prospects that exploit quantum correlations in electron microscopy.[116] A coincidence measurement setup based on entangled electron–photon pairs emitted from a vdW x-ray source, and realized in a TEM, is envisioned in Fig. 3(c).

IV. SHAPING FREE-ELECTRON WAVEPACKETS FOR CONTROLLABLE X-RAY GENERATION

The growing wealth of techniques to shape the spatiotemporal profile of an electron pulse provides undiscovered opportunities to control x-ray generation from these electrons. These techniques include both classical shaping of the electron charge and current distribution and quantum shaping of single electron wavepackets: using static fields,[117,118] radio frequency cavities,[119,120] laser pulses,[121–124] and material structures,[125] achieving spatial (temporal) shaping down to the picometer length- (attosecond time-) scale. The quantum wavepacket shaping of single electrons enables control over properties such as orbital angular momentum (OAM),[126,127] spin angular momentum,[128,129] and propagation trajectory.[130,131] Such capabilities have been achieved via breakthroughs in manipulating the phase-front of electron wavepackets[132,133] using amplitude and phase holograms,[134–136] nanoscale magnetic needles,[137] and electron–photon interactions.[138]

These advances in electron waveshaping techniques raise the fundamental question of whether quantum electrodynamical (QED) interactions (e.g., light emission) can be controlled via electron waveshaping. Schrödinger first interpreted the quantum wavefunction as the smooth charge density of a smeared-out particle.[139] Contradictions arising from

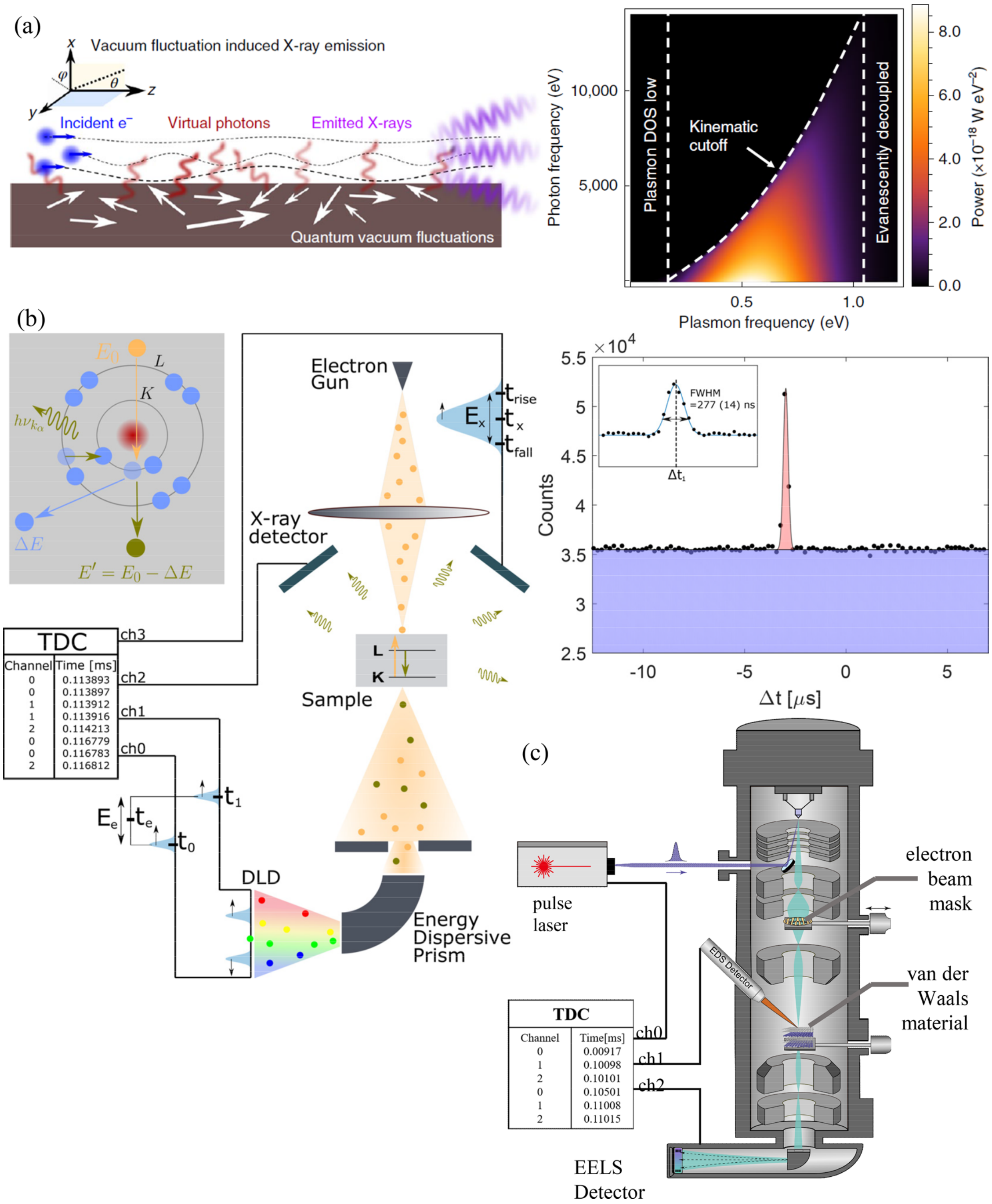

FIG. 3. Free-electron-driven x-ray quantum optics: emission of entangled photon–photon pairs (a) and electron–photon pairs (b) and (c). In (a), nanophotonic vacuum fluctuations give rise to a spontaneous two-photon emission process in which the outgoing infrared/optical plasmon and the outgoing x-ray photon are entangled (left). The diagram on the right shows the emitted photon power per unit photon frequency and plasmon frequency from an electron of velocity 0.99c traveling 5 nm above the surface of a graphene sheet, doped to a Fermi energy of 0.5 eV. In (b), collisional inner shell ionization (top left inset) is used to generate entangled electron–photon pairs used in coincidence measurements to significantly enhance the measured signal (left). The diagram on the right shows a histogram of the time difference between x-ray and electron energy loss (EEL) events showing a clear temporal correlation (red peak), with uncorrelated events present as background (blue). The width of the coincidence peak is shown in the inset. (c) A proposed setup for performing coincidence measurements using entangled electron–photon pairs, generated in this specific example via parametric x-ray radiation and/or coherent bremsstrhalung in van der Waals materials as per the scheme in Fig. 2(f). (a) Reprinted with permission from Rivera *et al.*, Nat. Phys. **15**, 1284–1289 (2019).[101] Copyright 2019 Springer Nature; (b) reproduced with permission from Jannis *et al.*, Appl. Phys. Lett. **114**, 143101 (2019).[115] Copyright 2019 AIP Publishing.

this view eventually led to the wavefunction being interpreted as the probability density of a point particle:[140] as Feynman put it, "the electron is either here, or there, or somewhere else, but wherever it is, it is a point charge." [141] On the one hand, this has led to an overall understanding that only higher order correlation measurements (rather than the intensity) would be affected by the wavefunction of the emitter.[142] On the other hand, it had been observed that an electron behaves exactly like a smooth charge density in stimulated emission processes, which have been shown to depend on the waveshape of the emitting electron in both experiment[143] and semiclassical theory.[144–146] This has led to much interest in the relation of emission intensity to the shape of the electron wavefunction.[147–152]

Through a fully quantum theory, we showed in a recent work that electron waveshaping can affect the emitted radiation[153] [Fig. 4(a)]. This surprising result occurs when different contributions to an emitted photon state are entangled to the same outgoing electron state. We applied our concept to bremsstrahlung, showing that it is possible to control this process of spontaneous emission from a free electron through the quantum interference resulting from electron waveshaping. Specifically, we show that free-electron waveshaping can be used to tailor both the spatial and the spectral distribution of the radiated photons, enhancing the directionality, monochromaticity, and versatility of photon emission compared to the unshaped case for both atomic and undulator bremsstrahlung. This theoretical concept agrees with the observations of an earlier work, where it was demonstrated theoretically and experimentally that using post-selection of the electron, spontaneous emission into near-field modes (rather than radiation) can depend on the symmetry of the initial electron wavefunction.[154]

Our study also revealed why such possibilities have not been seen before. For example, a recent experiment showed no dependence on

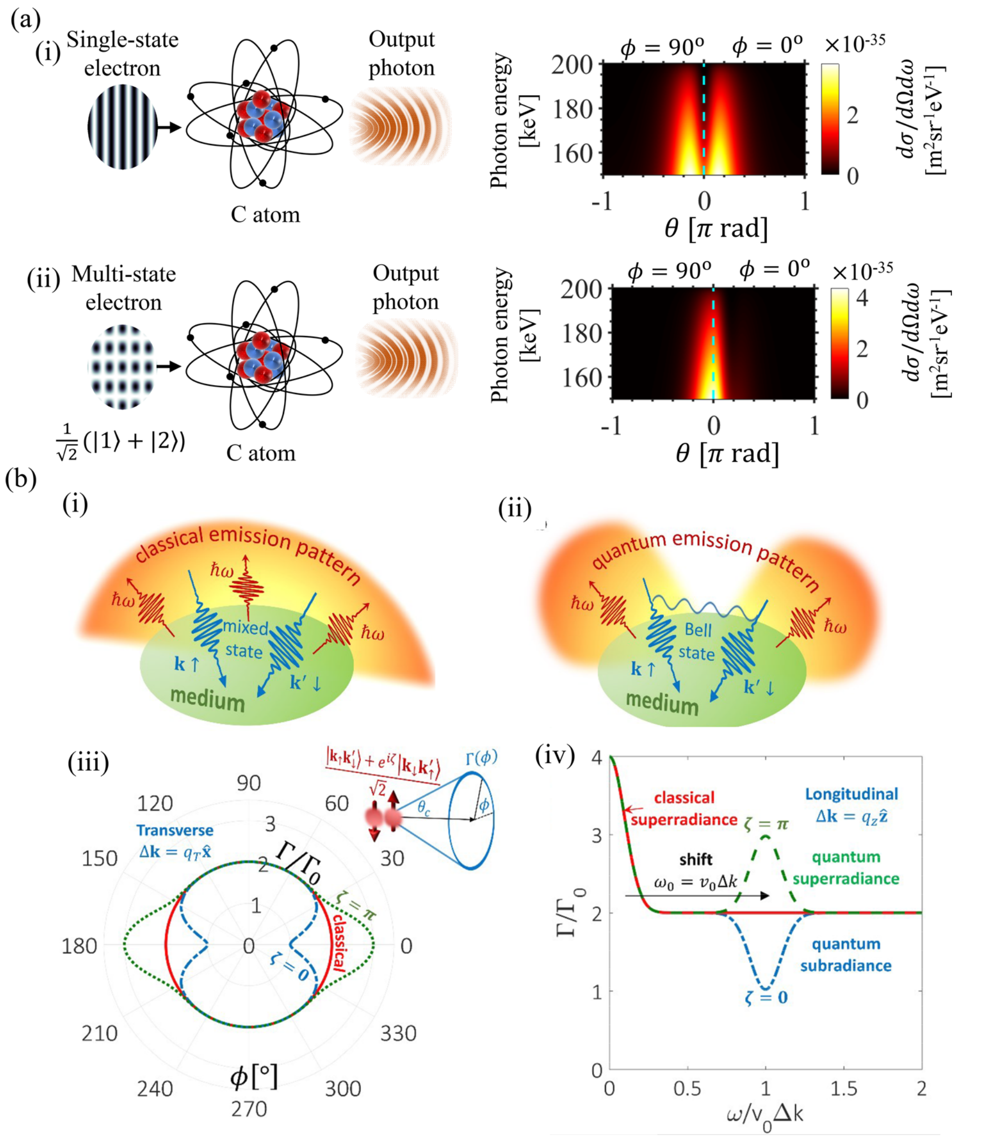

FIG. 4. Shaping free-electron quantum wavepackets for controlling radiation. (a) Shaping the single-electron wavefunction can be used to enhance the directionality of output x rays comparing the (i) unshaped and the (ii) shaped scenarios. (b) Multi-electron entangled states create radiation patterns that cannot be created from classical electrons. Illustration comparing the (i) classical (unentangled) and (ii) entangled scenarios. In particular, entangled electron states can create quantum superradiance, and subradiance is possible, significantly affecting the (iii) transverse profile and (iv) spectrum of the output radiation. (a) Used reproduced with permission from Wong *et al.*, Nat. Commun. **12**, 1700 (2021).[153] Copyright 2021 Authors, licensed under a Creative Commons Attribution (CC BY) license; (b) Figure reprinted with permission from Karnieli *et al.*, Phys. Rev. Lett. **127**, 060403 (2021).[149] Copyright 2021 American Physical Society.

wavefunction for Smith–Purcell radiation.[155] The reason for no wavefunction-dependence was that the contributions to the emission from different initial electron angles could not interfere because each photon state was entangled to a different outgoing electron state. Similarly, work that considered shaping Cherenkov radiation through the orbital angular momentum (OAM) of electrons[127] found no change to the power spectrum, unless the outgoing electron was post-selected. We attribute these spontaneous emission results to the electron behaving ultimately as a point-like particle (as nicely put by Feynman[141]) regardless of its wavefunction.

Combining electron waveshaping with free-electron nanophotonics concepts (Sec. II) suggests intriguing possibilities like tailoring the electron wavepacket periodicity to match with the atomic lattice of the nanomaterial. Significant modifications to the output radiation can also be achieved by using multiple entangled input electrons[149] [Fig. 4(b)]. The ability to tailor the spatiotemporal attributes of photon emission via quantum interference, thus, provides additional degrees of freedom for shaping x rays, creating exciting prospects for undiscovered x-ray physics. As a future prospect, electron beam shaping can potentially be used to suppress bremsstrahlung processes, leading to reduction in background noise and improving measurements of characteristic peaks. This would, in turn, improve contrast in x-ray spectroscopy techniques like x-ray resonance fluoroscopy.

V. OUTLOOK

X-ray sources based on free-electron nanophotonics are promising for applications that require high-brightness, dynamically tunable, monochromatic, and highly directional x rays. Using one of more of the directions above promises additional degrees of freedom for controlling different aspects of x-ray generation, even including the x-ray photon polarization. Traditional x-ray tubes fall short in these aspects due to lack of tunability—the output x-ray spectral peaks depend on the anode material instead of on electron energy, and the emission is isotropic and unpolarized.

In soft x-ray fluorescence spectroscopy, tunable energy excitation would allow one to investigate resonant phenomena and identify multi-electron excitations leading to x-ray satellite structures.[156] Furthermore, tunable x-ray photons enable site selectivity with respect to identical atomic species in different chemical environments since the x-ray photon energy can be chosen, so that a core electron is promoted to an unoccupied state with a strong localization on a particular site.[157] By leveraging the well-defined polarization of nanophotonic x-ray sources and detecting the x-ray emission with angular selectivity, one can obtain further information on bonding geometry, etc., of the sample.[158] These prospects are especially important in techniques such as ultrafast x-ray spectroscopy and phase contrast imaging that commonly take place in x-ray facilities and would benefit from a more compact and accessible source. It should be noted that phase contrast imaging has been achieved using conventional x-ray tubes with the use of spatially varying masks, for instance through Talbot-Lau interferometry[159,160] and edge illumination.[161,162] However, the presence of absorbing masks reduces x-ray flux, demands careful alignment and stability throughout a clinical examination, and requires precise fabrication of the masks, which are usually made of high-Z materials. These last two conditions are supposedly the most critical issues that have hampered the widespread use of these techniques in the clinical context.[163]

In free-electron-based x-ray generation processes where more than one electron is involved, coherence in the output x-ray radiation can be achieved via the structuring of the electron distribution. For instance, in x-ray free-electron lasers, the electron bunch co-propagates and interacts with the emitted x rays over long distances, leading to self-structuring of the electron bunch into sub-wavelength nanobunches. These subwavelength structures in the electron density are directly responsible for x-ray intensities that scale with N^2 instead of N[164] (N being the number of electrons). This process is known as self-amplified spontaneous emission (SASE).

In addition to self-structuring techniques, the electron wavepacket can also be structured by external processes[117–124] before being made to radiate x rays. A coherent, high-brightness x-ray source is potentially useful for many phase-dependent imaging techniques, such as coherent diffraction imaging and ptychography.[165] A recent work has shown that nanophotonic structures can be used to realize lasers based on stimulated emission by free electrons, in a process analogous to SASE, but on the nanoscale.[166] The associated threshold beam currents are in the nanoampere range and could be realized in electron microscopes. However, the work considers emission at infrared wavelengths, raising the exciting question of whether the scheme can be scaled to x-ray photon energies.

In conclusion, we have pointed out innovative research directions in x-ray science that open up unique opportunities for future x-ray technology and recently became more accessible due to advances in nanomaterials and quantum optics. We have described how the highly nonlinear and spectrally broadband nature of the free electron allows us to leverage the versatility of nanophotonics for the manipulation and enhancement of x-ray emission. We have identified three main themes that we believe to be of special interest: the study of emerging materials to serve as nanophotonic platforms for compact, tunable, and high-brightness x-ray generation; the quantum shaping of electron wavefunctions to control the spatiotemporal profile of emitted x rays; and the entanglement of output electrons and photons for coincidence measurement schemes. We hope that this Perspective has helped highlight the prospects for future research and will help drive future discoveries in the field.

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DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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