A perspective on novel sources of ultrashort electron and X-ray pulses

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\section*{Abstract}

Recently, much attention has been devoted to the development of new pulsed sources of radiation for investigating matter with atomic scale temporal and spatial resolution. While much has been achieved thanks to modern ultrafast laser technology, the ultimate coherent light source, the X-ray free electron laser (X-FEL), promises to deliver the highest X-ray photon flux in the shortest pulses at energies unreachable by conventional solid-state lasers. In parallel, other approaches that utilize electrons in tabletop setups as a probe have been developed demonstrating the potential for a valid complement to X-ray based techniques. Here, we consider yet another possible avenue in which the technology of femtosecond high energy electron microscope and propose a hybrid experiment with relativistic electrons as a probe and fs X-ray pulses as a pump taking advantage of both technologies.

\section*{1. Introduction}

In the temporal range between fs and ps, several relevant phenomena like rotations, vibrations and phase transitions in liquids, proteins and solids, are found. In Fig. 1 of Ref. [1], these timescales and the corresponding processes were shown. In summary, the processes that happen in this time-domain govern the functionality and properties of materials of fundamental, technological, and biological interest; in complex systems, their identification via spectroscopy becomes difficult and most importantly highly model dependent. For this reason, the possibility to directly image matter with temporal resolution in real space is of crucial importance. Overall, the new tools need to provide fs temporal resolution in time, approximately 1 Å resolution in space, and sensitivity to different states of matter like liquids and solids, often composed of light elements like H, C, O, N especially in biologically relevant systems. In complex structures like proteins, a high degree of spatial coherence is also needed for imaging large areas; more in general, for systems of biological relevance radiation damage should also be controlled. While the discussion of current techniques for studying structural dynamics is beyond the scope of this perspective and has already been extensively done [1], we intend to briefly compare those using X-ray photons to those using electrons with the intent to show how these can be complementary, and in future perspective even combined. In what follows, we will briefly review the quantitative comparison between different techniques, with a focus on the perspective of a new tool based on relativistic electrons. For the rest of the article we will refer to relativistic electrons for those with energies higher than 1 MeV for convenience reasons. This distinction is necessary when dealing with ultrashort electron bunches as above those energies space charge effects can be neglected allowing for shorter pulses containing more charge. The aim of this perspective is to simplify the subject into few key-concepts in order to provide an understandable, direct and somewhat blunt overview of what can be obtained with different approaches.

\section*{2. Comparison between different approaches}

A direct comparison between X-rays and electron based techniques is provided in Fig. 1. Different parameters are taken into account. In more details:

- **Pulse duration:** X-ray fs experiments at synchrotrons have been performed via slicing techniques [2,3]; this results in a severely reduced number of photons per pulse (roughly 10 to 200 photons in 100 fs depending on the energy bandwidth of the beam) [2,3]. Instead, modern X-FEL technology is capable of concentrating an extraordinarily high number of photons into less than a few fs [4–6]. In simple 30 kV electron diffraction set-ups in reflection geometry, pulse duration is limited by the space-charge effect. Controlling the propagation distance and keeping a low number of particles in the bunches have guaranteed temporal resolutions between 200 fs and 1 ps in stroboscopic experiments [7–9], and spot sizes between 6 μm [9] and 100–200 μm. Newer approaches have improved performances using pulses...
containing single electrons [10], or using higher energy electrons, 100 kV, and RF technology for compression resulting in sub 100 fs pulses containing up to 10⁶ particles per bunch [11]. Single shot relativistic ($E_{\text{kin}} > 1$ MV) electron diffraction experiments have been also performed streaking pulses containing 10⁸ particles [12,13]. The latter demonstrate the ability of RF technology to provide ultrashort electron bunches with substantially high charge, see Fig. 2. New projects are also starting combining RF compression technology with relativistic beams, and claim to be able to deliver relativistic electron diffraction with sub-10 fs pulses [14]. A new approach for compressing electron pulses based on the exploitation of the ponderomotive force has also been proposed [15] with the target of reaching the attosecond regime ($10^{-18}$ s).

- **Particles per pulse and total cross section:** In modern FELs, $10^{13}$ photons in fs pulses has been achieved [16]. As far as electron sources are concerned, 10–50 kV electron diffraction set-ups operate in the regime of $10^2$–$10^4$ particles per bunch. This allows stroboscopic experiments and a rather simple implementation, but it is certainly not able to provide single shot diffraction patterns. Recently, RF compression technology has been used to squeeze $10^6$ electrons in less than 100 fs pulses obtaining successfully single shot diffraction patterns at 100 kV [11], see Fig. 5 (e). To be noted at this point is the fact that, for a 200 nm thick sample, $10^6$ electrons are enough for a single shot diffraction pattern instead of the $10^{12}$ particles needed for X-rays as a consequence of the larger cross section of electrons for interaction with matter. At higher acceleration voltages, the cross section for electrons diminishes a bit (see Fig. 1) but space charge becomes negligible and $10^8$ particles per bunch have been obtained in Ref. [12]; with these many electrons, true single shot and time-resolved experiments are possible via streaking, see Fig. 2 (c). This technology allows not only to obtain a single shot image of a material, but also its dynamics across an irreversible phase transformation such as melting [12]. In general, in order to properly compare the sensitivity to materials one should look at the product of the cross section times the number of probe particles that can be packed in every pulse.

- **Coherence length:** The discovery of lasers made available optical light sources with unprecedented coherence properties, both spatial and temporal. Nowadays, FEL sources provide high intensity, almost fully coherent beams at X-rays energies, with huge coherence lengths that can be in the several microns range [17]. Full transverse coherence can be reached in seeded sources [18], while for hard X-rays SASE machines, a coherence length of approximately 300 μm can be obtained in a beam of 2.1 mm [19], corresponding to a degree of transverse coherence around 0.2 (obtained as the coherence length/beamsize).}

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**Fig. 1.** Comparison of the characteristic properties of ultrashort hard X-ray and electron sources, in relation to their performances in diffraction and imaging.

**Fig. 2.** Schematics of the relativistic electron diffraction set-up in [12] with an example of a single shot streaking experiment on a gold sample. In the left part, a representation of the MeV beamline at UCLA is displayed. The electron beam diffracted from the sample passes through an RF streak camera and in the right part of the figure one can see the streaked Bragg spots with and without laser excitation. In this configuration, the temporal evolution of the intensity decrease of the diffraction features induced by light excitation can be obtained in one single shot.
the longitudinal or temporal coherence length $L_T = \frac{\lambda}{\Delta E/E}$. Brightness $B \propto J L^2_L$ is a conserved quantity, so in principle coherence lengths cannot be discussed independently from the pulse duration and the number of particles per pulse. The degree of coherence required depends on the application: for crystal diffraction the coherence lengths should be at least of the order of the lattice spacing, i.e. ranging from sub-nm for simple atomic systems to tens of nm for proteins; for coherent diffractive imaging or holography the coherence lengths should be comparable to the sample size, which can be several microns. In principle any degree of coherence can be achieved by simply blowing up the beam, both longitudinally and transversely, but this goes at the expense of temporal resolution and the number of particles interacting with the sample. If one wants to retain the shortest pulse duration the temporal coherence length is basically fixed to a value of, typically, $10^2$–$10^4 \lambda$, i.e. $L_T = 10$–$100$ nm for electrons and $L_T = 0.1$–$1$ µm for X-rays. Clearly, temporal coherence is not limiting for even the most demanding crystal diffraction applications. It could be an issue in case of coherent diffractive imaging or holography applications, which lies however outside the scope of this overview and will not be discussed any further. In the remainder the discussion will therefore be restricted to spatial coherence. To enable few or single-shot operation, no particles should be waisted, so the beam diameter should be matched to the sample size. Since the spatial coherence length $L_s$ is proportional to the beam diameter $D$, $L_s$ that can be achieved depends on the size of the crystal sample. The proper figure of merit for spatial coherence is therefore the relative spatial coherence length $L_s/D$, which is proportional to the transverse emittance. In Fig. 1 we give the degree of spatial coherence in terms of the relative number $L_s/D$. For example, a 10 µm beam would have a coherence length of 1 µm for the X-FEL and only 10 Å for a 30 kV electron beam. In the case of electron sources, the charge of the beam is also specified in the table as the coherence length depends on this parameter. This being said, it is important to discuss what level of coherence is actually needed and for which scientific problem. Pulsed electron guns for kV electrons have been estimated to have beams with a coherence length in the order of $3$–$8$ Å [20]. This value is enough for crystallographic studies, as typical interatomic distances are in that order of magnitude; however, even in systems with much longer superstructures 30 kV electron diffraction delivers good quality patterns. In Fig. 3, a comparison between the diffraction pattern of a 2.7 nm superstructure taken with 30 kV electrons (Fig. 3(a), (b), (d) and (e)), 200 kV electrons (Fig. 3(c)), and X-rays is shown (Fig. 3(f)). The satellite streaks and peaks of the modulation of a BSCO crystal [21,22] (Fig. 3(b) and (e)) are clearly visible in all images in spite of the fact that in theory the coherence length of 30 kV electron beams should be almost one order of magnitude too small. At higher energies, in the 100s kV regime, better coherence properties are obtained in transmission electron microscopes (TEMs). Pulsed relativistic electron sources potentially provide better coherence.

![Fig. 3. Bi2Sr2CaCu2O electron and X-ray diffraction. (a), (b), (d) and (e) Diffraction patterns in RHEED geometry of a Bi2212 sample [21,22] taken with 30 kV electrons with estimated coherence length of 5 Å. The colored arrows in panels (a) and (b) represent the direction in which cuts in panels (d) and (e) are displayed. The superstructure modulation of 2.7 nm and the c-axis lattice parameter of 3 nm are both resolved. (f) Transmission through a 200 nm Bi2212 film obtained in a 200 kV TEM showing evidence the satellites of the 2.7 nm superstructure. (f) BSCO crystal and the 2.7 nm super structure. (g) X-ray diffraction pattern showing the same satellites of the super structure.]()
properties as well because of the high extraction field at the cathode which allows a smaller emission area. In addition space-charge effects are relativistically suppressed [23]. As far as diffraction from large biological samples like proteins is concerned, in which coherence is an important parameter, the first structural refinement of the protein bacteriorhodopsin was reported by TEM [24,25], (see Fig. 4 (a) and (b)). A sequence of diffraction images had been analyzed and allowed retrieving the structure of the protein identifying the proton transfer channel and all the functional substructures. The size of a protein can range from few nm to several tens of nm. However, the substructures contributing to the diffraction patterns have obviously smaller size, in the range of the coherence length of electron based instruments like TEMs. Therefore, for membrane proteins (which are the majority) electron diffraction and microscopy seems to be the most reasonable choice [26]. Overall, for protein diffraction a coherence length in the order of tens of nm seems to be sufficient. This can be easily achieved in static TEMs, and according to our estimates is also feasible with hundreds kV to MV pulsed sources with about 10^6 electron per bunch. The huge coherence length of X-FEL sources can be exploited for example in the study of protein nanocrystals [27]; in Fig. 4 (c) and (d), an image of nanocrystals is shown and their X-ray diffraction pattern. Each Bragg spot is split into several satellites resulting from the nanostructures and visible thanks to the long coherence length of the X-ray beam.

- **Spatial resolution:** In optics, spatial resolution is ultimately limited by the wavelength of the photons. For the X-ray sources considered here we can quote a resolution of the order of 10 Å. The associated wavelength of electrons is normally smaller than that of X-rays, resulting in a higher spatial resolution, in the order of 0.5 Å at 1 MV [23].

- **Particles per image:** Here, we want to provide a rough estimate of the number of events needed to produce an image of the same sample with X-rays and electrons. This quantity is important for addressing the radiation damage produced in biological specimens. It is instead of less concern for the study of solids and thin films. We take as a reference sample a cystine protein which is one of the least resistant specimens to radiation [28,29]; a dose of 10^7 Gy is already sufficient to damage this protein. In the experimental conditions of [29], this level of absorbed radiation can be obtained by exposure to 10^{12} X-ray photons, which is close to the exposure needed for obtaining an image with 2 Å spatial resolution [29]. As far as electrons are concerned, 10^7 Gy correspond to an exposure to a beam delivering 5 e^-/Å^2 at 1 MV [28]. In order to obtain an image with

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**Fig. 4.** Protein imaging with electrons and X-rays. (a) and (b), The first protein structure retrieved from a bacteriorhodopsin sample in a TEM, diffraction and the real space corresponding model, respectively. In panel (a) The vertical lines are individual lattice lines. Their shading is modulated according to their diffraction intensity. These figures and descriptions are taken from [25]. (c) and (d) Protein nanocrystals and their coherent X-ray diffraction pattern [27].
high energy electrons, roughly 10 electrons per pixel are needed, see Fig. 5 (c) [30]. In order to obtain the same resolution of 2 Å quoted for the X-ray data on a field of view of $60 \times 60$ nm$^2$, an image containing $5 \times 10^3$ pixels is needed, therefore $5 \times 10^6$ electrons need to irradiate the field of view. This results in an exposure in the order of $5 e^-/\AA^2$, just enough to produce an image and damage the cystine protein. In this respect, high energy electrons and X-rays seem to deliver very similar performances, while the situation may be slightly better with lower energy electrons, however at the cost of coherence length and spatial resolution. Recently, it has been proposed that thanks to the possibility of squeezing a sufficient number of photons in ultrashort, few fs X-ray pulses, the protein could be imaged before the actual damage occurs. What we just showed is that the very same approach would apply to electron diffraction where one can concentrate the same “electron equivalent radiation dose” needed to damage a protein in a similar ultrashort pulse. It has to be noted that for relativistic electrons a significant source of damage comes from knock on processes; a careful discussion of the electrons damage as a function of the accelerating voltage is found in Ref. [28]. In general, despite the fact that the damaging mechanism between X-rays and electrons can be very different [28,29], a phenomenological estimate can be done in order to relate the dose received by a specimen and the quality of the images obtained by electrons and X-rays. In this respect, based on our estimates we conclude that similar radiation doses are needed to spoil imaging in both electrons and X-rays techniques. Also, it has to be noted that indeed protein crystallography has been performed successfully with hard-X-ray pulses shorter than the typical damage timescales [27,31].

- Penetration depth: Owing to their higher cross section for interaction with matter, electrons have shorter penetration depth than X-rays. This allows to study efficiently bi-dimensional systems, like membrane protein crystals [32]. Most proteins do not crystallize in a thick 3D sample and are therefore more suitable for electron based experiments [28]. However, a longer penetration depth is required for studying proteins in their natural environment, like in a liquid solution for example. In this case, $\mu$m penetration depths seem to be needed, which are easily obtained with X-rays [33]. In Fig. 1, the numbers quoted for the penetration depth refer to the interaction between radiation and solids. In this case, relativistic electrons can penetrate half a $\mu$m [34], but it may be even larger in liquids. When imaging thick specimens with electrons, a chromatic aberration corrector or an energy filter can be considered in order to avoid loss of contrast and image quality due to inelastic processes. The energy filter will simply remove the electrons that have lost energy, while a $C_z$ corrector will avoid blurring due to chromatic aberration; this could lower slightly the number of useful events. As a last remark on this subject, in a pump–probe experiment the pump and the probe beam ideally need to have similar penetration depth. At typical excitation energies (few eV) this condition is satisfied by a hybrid experiment pump-photons/ probe-electrons, while it is not satisfied when probing with X-rays. On the other hand, it would be satisfied if pumping in the THz regime, of great interest for materials science, and probing with X-rays [35].

3. Detection of high energy electrons

The issue of detectors is a very important one as what matters to the signal-to-noise ratio is not only how many electrons hit the sample, but how many diffracted electrons are captured by the acquisition system. In particular, the characteristics of the detector are high sensitivity and good spatial resolution, quantified respectively by quantum efficiency and point spread function. For nonrelativistic ultrafast electron diffusion (UED) there are well established methods and instruments to image keV electrons with high detection efficiency. Micro-channel plate (MCP) is a key element that is used to detect and amplify either the keV electrons or the low energy electrons inside an image-intensifier. The amplified electron flux is then converted by a scintillator to visible photons which are subsequently fiber-optically coupled to a high efficiency charge-coupled device (CCD) camera. Due to the large gain of the MCP and the high light collection efficiency of the fiber-optics coupling, it is relatively straightforward to achieve every-electron detection capability (quantum efficiency =1) with acceptable spatial resolution ($\leq 200 \mu$m); these are the typical performances obtained in studies like Ref. [8].

For relativistic (MeV) detectors, a better choice for the detection is the use of optimized passive fluorescent screens which are low cost and can provide high electron-to-photon conversion efficiency and good spatial resolution. A phosphor screen, taking advantage of the large penetration depth of MeV electrons, could yield as many as a few thousand photons for each MeV electron. The point spread function is usually limited by the thickness of the screen, but it can be kept well below $50 \mu$m. Very recently, [36], have demonstrated the use of a combination of fluorescent screen (P43 phosphor) lens coupling (F#1.4 or better) and gain camera (EMCCD or ICCD) to obtain single electron detection capability even at MeV energies.
4. Future developments of these tools

Triggered by the interesting developments of ultrafast electron technologies, more advanced and exotic strategies have been proposed/developed for pushing further the limits of these techniques. Relativistic electrons of 3 MeV energy have been shown to be suitable for single shot electron diffraction with sub-100 fs temporal resolution and pulses containing up to $10^8$ particles [12]. In order to push the limit of a relativistic electron based device, it is possible to apply the idea proposed in [11] taking advantage of the longitudinal focusing kick imparted to the beam by a properly phased radiofrequency accelerating cavity. If the cathode in a RF gun is illuminated by an ultrashort laser pulse, the beam undergoes a space-charge driven expansion which generates an almost ideal uniformly filled ellipsoidal distribution. Such distributions are advantageous since they are characterized by linear space charge fields and extremely low emittance longitudinal phase spaces, ensuring good compressibility [37,38]. In the scheme proposed at the UCLA Pegasus Laboratory it is planned to use a coupled cell 50 cm long accelerating cavity with a gradient of 20 MV/m to recompress the 4 MeV few pC beam out of the RF gun to a bunch length <5 fs (according to simulations) [39]. Optimization of this scheme could even result in sub-fs pulse lengths. In this context, the use of a collimating skimmer is important as it removes the problems associated with path length differences for the particles generated at the outer edge of the beam. This also allows to select the central part of the beam having the best emittance and hence to improve the coherence length. The charge in the relativistic beam can be further reduced given that recent results [14] show that by proper choice of fluorescent screens and CCD cameras, it is possible to obtain single electron detection capability even for relativistic electrons. This is important as it closes the gap with the non relativistic setups which had so far a significant advantage thanks to the high detection efficiency of microchannel plates coupled to image intensifiers. This approach therefore delivers very short pulses containing a large number of particles allowing single shot experiments with extreme temporal resolution. In addition, better coherence properties for high energy pulsed electron beams can also be obtained, approaching what is currently achievable in modern TEMs [23]. The detrimental effects of space charge forces can even be completely eliminated by proper shaping of the photo-emission laser pulse [37,38], making it possible to fully conserve the spatial coherence of the beam from photocathode to target, also at non-relativistic speeds. This is an approach similar to what was done in [39], but applied to the temporal coordinate. The next step is therefore to improve the coherence at the photocathode surface. This is usually done by reducing the emission area, a strategy which has been developed to high levels in TEMs, culminating in the use of a single carbon nanotube field-emitter [40]. For high-charge femtosecond operation, however, the size of the emitting area is limited by the available acceleration fields for metal cathodes, which have to overcome image charge fields associated with the instantaneous extraction of large bunch charges. The field strengths offered by state-of-the-art relativistic RF photoguns allow femtosecond photoemission of up to $\sim 10^3$ electrons per $\mu$m$^2$, an order of magnitude higher than in DC photoguns [37,38]. Therefore, room for improvement on the source design is available and performances better than the lower voltage beamlines are to be expected. Another possibility has been put forward for improving the coherence properties of pulsed electron beams, based on reducing the electron temperature of the source instead of the emission area [41]. By near-threshold photo-ionization of a cloud of laser-cooled Rb atoms a source with an effective electron temperature 2–3 orders of magnitude lower than conventional photo-emission sources has been realized, i.e. an improvement of the spatial coherence by more than an order of magnitude for the same emission area [42]. Combining such an ultracold electron source with RF acceleration techniques should enable the high bunch charge and beam coherence required for single-shot crystallography of proteins [43]. At reduced bunch charges the spatial coherence can be improved even further, eventually matching the beam quality of state-of-the-art TEMs. Interestingly, these sources also provide ultracold pulsed ion beams [44]. While the relatively slow ions are traveling towards the target, a synchronized electron bunch can be generated which overtakes the ion bunch. In this way electron–ion pump–probe experiments may be realized.

4.1. A fs 1 MV TEM

Provided the room for development of the approaches discussed above, it appears that a rather interesting direction could be the development of an “intermediate-scale” facility for imaging and diffraction. In particular, high energy TEM (1 MV and above, often called UHVEM for ultra high voltage electron microscopes) are still being built and installed. At such acceleration voltages, pulsed operation is favorable and the related advantages have already been discussed in Refs. [45–47]. Transmission electron microscopes at acceleration voltages higher than 0.4 or even 1 MV have initially been developed and flourished in the 70s as a way to improve the lateral resolution. Indeed, while the electron wavelength of 2.5 pm at 200 keV should in principle be sufficient for ultimate atomic resolution, the aberration of the lenses (spherical and chromatic) has been the main factor limiting resolution over decades. Reducing the focal length (by reducing the gap in which the sample is placed), reducing the energy spread of the gun and increasing the acceleration voltage were the only three possibilities to approach the 1 Å resolution limit [48]. While increased acceleration voltages is in principle only beneficial to resolution, in practice, it is followed by a list of practical drawbacks: mechanical instability of the huge system, problems with the heat generation in the lenses, stability of the high voltage among others. As a consequence, only few systems where built worldwide: the only commercially available UHVEM is currently the JEOL 1.25 MV TEM. Around the year 2000, two special systems were developed in Japan by Hitachi, a 3.5 MV machine, which is the TEM currently in operation with the highest voltage [49] and a 1 MV FEG instrument with a resolution of less than 0.5 Å [23]. This 1 MV FEG microscope has a very good coherence length, in the tens of nm regime, Fig. 6(b), (d) and (e).

Since correctors for the spherical aberration ($C_s$ correctors) and later chromatic aberration ($C_C$ correctors) have been made commercially available [50], the trends lie much more on the reduction of acceleration voltage. Lower voltages seem to have only advantages, the knock-on beam damage is lower and sensitive materials can be imaged for hours without being destroyed [51], Valence EELS suffers less of Cherenkov radiation [52], and the stability of the high voltage, lens current, etc., are easier to keep. In terms of diffraction analysis, it was shown that diffraction patterns acquired at very high voltages suffer more from dynamical diffraction effects [53] and the intensities of the diffracted beams are more difficult to interpret. Precission electron diffraction nowadays allows to cancel dynamical effects and produces therefore diffraction patterns that are easier to interpret even at conventional medium acceleration voltages. UHVEM is now reduced to very specific cases, namely for the investigation of very thick specimen that cannot be easily thinned by conventional methods (and with the use of focused ion beam for TEM sample preparation, this is less and less of an issue) or when the interest is the radiation damage of materials, as for example in the nuclear industry. As a consequence, $C_s$ correction has never been used on an UHVEM because
Cs correction and very high energy are generally considered as two antinomic approaches for improving spatial resolution. UHVEM however is extremely interesting for dynamical experiments as discussed above. The design of a modern setup for ultrafast experiment at very high voltage should take advantage of the recent developments in terms of aberration corrected microscopy. Computer controlled correctors are nowadays very stable and can be operated for at least a full day without requiring a tuning. In principle nothing prevents the design of a Cs/Cc corrector for a 1 MV TEM [54], and an ultrafast UHVEM could for example use an objective lens with very large gap, allowing for more space for the laser setup and in-situ experiments (e.g. injection of molecules for single molecular analysis). One could even think of a design using a normal projective lens as objective lens, as recently proposed for a 100 kV microscope [48]. In this configuration, even without Cs/Cc corrector, the resolution might be slightly degraded but the gain would be enormous since the sample could be handled in a large chamber free from magnetic fields allowing the study of magnetic phenomena. Also, it is important to notice that lower energy electrons have smaller penetration depths and produce more inelastic radiation damage, particularly critical for organic samples [28].

In this configuration, single shot fs electron microscopy could also be feasible, since it has been already demonstrated in the ns regime [55,56], and provided a relativistic source of shorter bunches is feasible. At these energies, a very high contrast to magnetic features is also available, making Lorentz microscopy optimal [34]. Fig. 6 shows a typical image of a gold film taken with 0.5 Å resolution. In Fig. 6(c), we also show an example of the image of a vortex lattice in high temperature superconductors, where individual vortices are very clearly observed at low field. This extraordinary contrast for subtle magnetic structures may be exploited in combination with ultrafast technology for studies of fundamental magnetic properties of materials, as well as for studying light-induced spin cross-overs in biomolecules [2]. Thanks to the high cross section of electrons, the relatively higher coherence length in the MV regime, the possibility to squeeze more than $10^7$ particles in fs pulses, and a longer penetration depth up to half a μm in the bulk, this tool may also provide the ability to perform single-shot protein crystallography in different environmental conditions.

4.2. X-rays and electrons complementarity

In this paragraph, we wish to summarize what can be done with X-rays that cannot be done with electrons techniques and vice versa. As we discussed above, the main advantage of photon sources over current electron ones is coherence, and the possibility to perform high energy-resolution spectroscopy (the equivalent electron energy loss spectroscopy with relativistic beams will not have a resolution better than 1 eV [57]). For this reason experiments like protein nano-diffraction [27], or more generally holographic experiments will be more suitable with X-FEL sources. Also, the investigation of radiation damage due to exposure to intense ionizing radiation is obviously meant for high brilliance X-ray sources. The study of matter under extreme conditions such as high pressure, which requires the use of pressure cells, or high magnetic field, are also suitable for X-rays experiments as electrons cannot go through closed environmental cells, nor do they like intense magnetic fields. On the other hand, the study of time-resolved radiation damage due to charged particles irradiation is of course feasible with electron-based tools, and other effects such as the photon induced near field microscopy for imaging transient fields at interfaces and membranes [58], high energy Compton scattering for valence band spectroscopy [59], and Kikuchi nano-diffraction [60] are unique to electron probes.
4.3. FEL-TEM pump probe experiments

Another very interesting turn of the technologies discussed in this perspective would be to combine fs X-ray pulses with relativistic electron microscopy. In fact, X-rays are not only a unique tool for probing materials and molecules, they also provide the possibility to photo-dope materials with chemical selectivity. In a pump-probe experiment, the role of the pump pulses is crucial for determining the microscopic path through which photoexcitation evolves [61,62]. Typically, ultrashort laser pulses in the visible light region are used because of their easy availability. In this energy range, temperature jumps, charge transfers, and phase transitions can be excited. However, at these energies in many systems the strong orbital hybridization makes chemical selectivity of the pump absent, and in some cases like strongly correlated solids, it is even impossible to exactly decipher what light excitation is inducing in the sample since a thorough description of the low energy optical absorption can be lacking or be model dependent [22]. Very novel and interesting experiments have been put forward where a more precise selectivity of the excitation has been obtained via the use of THz pump pulses [63]. In the low energy region of the infrared spectrum, light can excite selectively atomic motions, providing a way to distort “ad hoc” the structure of a system for probing its consequent dynamics. This approach takes advantage of the interaction between light and low-energy many-body bosonic objects. Because of the way THz pulses are generated and their intrinsic characteristics, sub-ps temporal resolution is challenging in this case.

Another possibility would be to use X-ray pulses tuned to specific core levels for photodoping materials and studying their fs dynamics. An X-ray tuned to an element core transition will induce an extra electron in the conduction band of the material, leaving a deep core hole behind. Such a process is chemically selective, and easier to model since the character of the photoinduced carriers is known via dipole selection rules and matrix elements. Despite the short core-hole lifetime (few fs), it has been shown that chemically selective photodoping can happen in materials through cascade decay processes of the electron hole pairs, or by directly promoting itinerant carriers in valence band orbitals [64]. In order to clarify this point, we show an example of an experiment based on this idea: in Fig. 7, we show the in-plane Cu3d_{x^2-y^2} orbital hybridized with the oxygen 2p in a high temperature superconductor. It is well known that the carriers responsible for superconductivity can be doped by extra oxygen added to the specimen. In Fig. 7 top right panel, the absorption spectrum of the oxygen K-edge is displayed for different angles of the polarization of the incoming X-rays. A strong pre-peak absorption feature is visible around 523 eV, with a polarization dependence that reflects the shape of the Cu3d_{x^2-y^2} orbital. This spectroscopic feature has been assigned to the excitation of an oxygen 1s electron, and its promotion into the Cu3d_{x^2-y^2} orbital hybridized with the O 2p in the plane [65]. In a hybrid X-ray pump, electron probe experiment with a relativistic TEM, fs X-ray pulses may be used for photodoping a charge transfer insulator inducing superconductivity [64], and Lorentz microscopy may be used for probing directly the Cooper pairs superfluid properties via imaging, as shown in the bottom panel of Fig. 7. At the same time, structural motions could also be observed in diffraction. Such an experiment would deliver fascinating information on the mechanism of Cooper pairs formation in superconductors, but very similar concepts may be applied to a variety of problems where chemically sensitive photoexcitation can induce phenomena like magnetism, ferroelectricity, multiferricity or functionality changes in molecules.

Fig. 7. Top panel: The absorption of X-rays tuned with the oxygen K-edge in cuprates reveals a pre-peak feature, red circle, whose symmetry follows that of doped holes in the d_{x^2-y^2} orbitals in the Cu-O plane; when the electric field of the exciting light lies perpendicular to the plane, the pre-edge feature vanishes, as it also does changing doping level [65]. Pumping a cuprate at this wavelength could populate more selectively low energy excited states through resonant decays. In the bottom panel, we show a typical image of the vortex lattice in BSCCO obtained by Lorentz microscopy [34]. The dynamics of superconductivity upon photo-doping could be followed in this experiment.

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References

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