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New Technologies for X-ray Standards and Applications

A study report

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New Technologies for X-ray Standards and Applications
A study for the NMS Quantum Metrology Programme

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ABSTRACT

Very rapid progress has been made over the last decade in x-ray technology. The brightness and coherence of available sources has increased remarkably. This report is concerned particularly with, the development of so-called ‘table-top’ x-ray laser sources, the generation of high order harmonics of intense ultrashort optical pulses and advances in soft x-ray optics. These technologies combined open up new areas of metrology and research, they also provide the dramatic scaling down of experiments that once required accelerator-sized sources and hence make this work more widely accessible to industry and small-scale science. This may well lead to much wider adoption of x-ray techniques particularly those using coherent x-rays.

More speculatively, the potential of interactions with nuclear energy levels is discussed. Mössbauer levels have high energies and very high quality factors, and hence are potentially useful as x-ray frequency references; the realisation of this will require further dramatic advances in x-ray technology and metrological techniques.

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1 Introduction

This review covers a number of areas of current technological development grouped under the banner title x-ray technologies. It concentrates on what have become known as table-top x-ray sources, a technology for x-ray production and control that in some circumstances rivals large-scale accelerator facilities. The review is a survey of the status and prospects of work underway globally, with a view to considering advances in metrology that may be achieved or required by this rapidly advancing technology.

It is limited in scope to a number of areas of current research in x-ray technology with potential to be relevant to advancing metrology. Some other areas of x-ray production and application are also considered for background information, but the field is far too large to be considered in its entirety in a study of this scale. A number of the references cited are reviews which comprehensively cover an area summarised in this report, and where this is the case, that is noted with the citation.

The applications of x-rays are extremely diverse and some principal ones are listed in the review. Metrological applications are likely to be in short length and high frequency metrology in support of the other applications. Important x-ray applications now and in the future are the production and measurement of nanoscale features, principally for electronic chip production; materials structural analysis and biological, chemical and cell structure analysis. Other applications of growing importance are induction of inducing tailored chemical reactions and materials analysis.

New x-ray sources will require and test the physics of plasmas and highly excited atomic systems.

The current methods of x-ray production are outlined. Then we describe new methods developed over the last decade that are in some sense rivals to the large-scale facilities that have been used hereto.

These new technologies are based on a few critical advances; capillary phase matching, which allows high harmonic generation, Kerr-effect phase locking, which gives femtosecond length pulses, allowing high field levels without needing high average power. They have led to rapid progress in x-ray and ultrafast technology and in particular to the development of table-top x-ray lasers the size of an optical table rather than an accelerator facility. The large scale facilities have also been advancing and can now generate much higher brightness than earlier. Facilities now being planned will give a further four orders of magnitude increase in brightness in next generation devices.

2 Applications

X-rays were being put to practical use within a few weeks of their discovery in 1895 and within a year were an established medical tool. They are now used in a wide number of areas of physics chemistry and biology as well as medicine and have direct metrological use. A number of current and potential applications are reviewed in this section. I have not attempted to be exhaustive in this as the range of use of x-rays is astonishingly wide but I have tried to at least mention major areas and expand on some of particular relevance. The details of technological advances are not included in this section but are discussed further in section 4.

2.1 Metrology

The production of very short pulse lengths necessitates the use of high frequency radiation to allow a few cycles of phase in the time that the pulse lasts. As well as physics applications, the recent advances in the production of attosecond pulses could have important applications in improving timing and synchronisation capabilities.

Nanometer scale length metrology will require a ruler of appropriate wavelength. Use of the silicon lattice spacing is difficult and not accurate in comparison with the accuracies achieved at longer lengths using optical interferometry

X-ray frequency metrology could be brought up to the high accuracy achieved in lower energy regions. Giving extended wavelength coverage by precision frequency metrology. Potential future x-ray standards could be based on hydrogenic systems where electron transitions are in the x-ray regime or on Mössbauer transitions where nuclear transitions are used. To make the comparisons either an extension of the femtosecond comb technology could be envisaged perhaps by using high harmonic generation processes. An alternative method may be to make use of calculable systems such as He^+ where transition frequencies can be calculated from first principles in terms of fundamental constants.

The adoption of high frequency techniques for the applications listed below will require improvement in the techniques for characterisation and measurement of fast pulses. In recent years the measurement of the phase as well as the intensity of pulses has been developed using techniques such as frequency-resolved optical gating (FROG) measurement, which is now a well-developed system. [1]

2.2 Others

2.2.1 Lithography

Semiconductor chip fabrication is a huge industry and a major driver in the production and measurement of small features. Scale sizes of semiconductors continue to shrink rapidly so the size of features to be fabricated and measured grows ever smaller. Current fabrication technology is at the sub micron level and the next generation of chip production will be at 90 nm length scale. For optical lithography to continue downward in scale then higher frequency sources will be needed. The next stage of development will be into the EUV range; with a wavelength of 13.4 nm being suggested, as there are high reflectivity multilayer mirrors in MoSi

available at that wavelength[2]. This will be an extremely demanding application for all aspects of optical technology including metrology[3].

2.2.2 Biological applications and the ‘water window’

Molecular structure determination is a field that has relied on x-ray sources to make the strides that it has shown over recent decades. It has led to a revolution in the life sciences, and has the potential for further great advances enabled by higher performance sources. Ultra-bright x-ray sources may allow the determination of protein structure without the need to produce crystals of the protein. The new advanced x-ray free electron lasers or the capillary lasers, both described below, will allow tens of or perhaps single biomolecules to be analysed. This is particularly advantageous as some features of a protein’s structure and function will only be apparent when it is in solution; current x-ray techniques require the protein to be frozen in crystalline form. Some biologically important proteins such as membrane proteins are hard to crystallize and hence there is currently little good structural data on them[4].

Vibrating molecules can change shape on a femtosecond timescale and hence the production of femtosecond pulses would open up these vibrations for study, giving real-time information on internal molecular dynamics in addition to structure.

A particularly useful wavelength range for biological studies is the ‘water window’. This is the range between the K-absorption edges of oxygen (2.3 nm) and carbon (4.4 nm). In this window x-rays highlight the difference between organic matter and water and so it is especially useful for investigating organic specimens.

A working x-ray microscope has been demonstrated using a x-ray laser source. It has been used to image live hydrated organisms giving high resolution information on molecule and cell structure. The goal of this work is to produce x-ray holograms of cells initially for holographic studies of virus structure. X-ray microscopy potentially offers atomic resolution using the next generation x-ray sources such as those described in section 3.2.2. The principle of holography was developed by Gabor as an attempt to improve the electron microscope. It is only subsequently that it has been thought of as necessarily optical laser based.

2.2.3 Medicine

The earliest application of x-rays was as a diagnostic tool in medicine and this is still done both in a similar way to the earliest shadow photographs and using much more sophisticated techniques such as computerised tomography and the use of contrast agents.

2.2.4 Chemical applications

Carefully-shaped femtosecond pulses of light have been demonstrated to enhance the yield of chemical reactions and synthesize novel molecules. They are particularly effective in the synthesis of complex molecules. This has great potential for the chemical and pharmaceutical industries where the tailoring of reactions by catalysis is already a vital part of chemical synthesis, but efficiency of conventional techniques is often poor. The techniques used are discussed in section 4.2.

2.2.5 Atomic Physics

The availability of x-ray sources bootstraps the x-ray research itself. For example, the study of inner shell dynamics will lead to advances in atomic physics and understanding of plasma laser interactions. This will in turn lead to improvements in the x-ray laser sources as well as techniques for plasma diagnosis. The electron beam ion trap (EBIT) was developed to provide information on the atomic physics of highly charged ions as part of the x-ray laser programme. It now plays a part in the testing of the fundamental predictions of quantum electrodynamics.

2.2.6 Solid State Physics and Surface Science

Because of the match between interatomic distances and x-ray wavelengths, x-ray diffraction is an invaluable tool for probing the atomic arrangement in the solid state, both in bulk crystalline material and on surfaces. A huge amount of data on materials properties and surface properties has been acquired using synchrotron sources of x-rays.

2.2.7 Communications

Ultrashort electrical and optical pulses are used in communications and the pressure to increase data transfer rates means that there will be a continuing need to work with ever shorter pulses. This will inevitably require the use of higher frequencies.

Line of sight laser communications are being developed. Their principal advantages are high data rates and the extra security offered by directional signals. These may be developed at higher frequencies but would be restricted to lower wavelengths in the atmosphere because of the reduced transparency in the ultraviolet and perhaps by atmospheric distortion, although distortion can be mitigated by the use of adaptive optics. The absorption by the atmosphere that is strong in the ultraviolet is reduced again in the x-ray region so at greatly increased frequency this application again looks attractive. Use in space for inter-satellite communication would be unrestricted by atmospheric effects.

3 'Traditional' X-ray sources

3.1 Difficulties of x-ray production and control

Progress in x-ray devices and applications has not been as rapid as that made in visible optics because of a number of practical problems.

The standard tools of optics, simple lenses and mirrors, as well as complex ones such as optical modulators, become harder to produce as wavelengths become shorter. This is because of material properties changing with wavelength. In the x-ray regime beyond about 50 nm all materials have a refractive index close to one so that using multilayer structures for mirrors or filters is not possible. As wavelengths become smaller the size and finish of components that must match the wavelength become rapidly more difficult to manufacture.

A monochromatic x-ray source avoids the problem that x-ray optics tend to suffer from chromatic aberration and recent advances in soft x-ray optics[5] have also improved the utility of x-ray sources.

For laser action the gain conditions that must be met for laser action to occur become rapidly more difficult as the wavelength shortens. The spontaneous emission coefficient scales as roughly λ^{-2} and the pump power to maintain a given population inversion as λ^{-3} . Thus the power density required to maintain a given gain scales as λ^{-5} therefore an x-ray laser will require in the order of 10^7 to 10^{12} times more pump power density than an optical one[6]. Cavity enhancement becomes ineffective as the reflectivity of mirrors is reduced with increasing frequency. Limited gain and very high pump power requirement have been the main problems in the development of x-ray laser.

The behaviour of the source is complex particularly because the laser or harmonic generation material is a plasma. Atomic physics data and plasma modelling is needed, one source of which is the electron-beam ion trap, the UK facility is based in at Oxford with others in the USA and Japan.

The status of the recently developed or proposed x-ray sources was reviewed in Science magazine in 2002[7]. The article was billed as a 'battle' to become the next generation source. The table-top sources were seen as an alternative to accelerator sources in some applications. It is most likely that both will find their niche.

3.2 Accelerator based sources

Electrons subject to acceleration will radiate electromagnetic radiation. In an antenna where the electrons travel in wires the acceleration is very weak and the radiation is in the radiofrequency range, in circular high energy accelerators where the energy range can be in GeV the acceleration is much greater and hence the radiation produced as the electrons are accelerated around the ring can extend from millimetre waves to hard x-rays. This radiation is known as synchrotron radiation. Synchrotron radiation was first seen in 1947 but thought of as a problem that reduced the energy of particles in synchrotrons designed to produce high energy collisions. However, by the 1960s it was realised that synchrotron radiation was a useful source with unique properties;

so ‘second generation’ synchrotron sources were built as light sources rather than particle accelerators.

In some cases, rather than using the unavoidable bending provided by the ring, ‘wiggler’ or undulator sections are incorporated consisting of sections of bending magnets with rapidly reversing polarity that can increase the acceleration by an order of magnitude.

Synchrotron radiation has a number of outstanding properties; continuum over very high wavelength range, high intensity, highly collimated, highly polarised, high brilliance. The high cost of these sources has been justified by their finding a wealth of uses particularly in determining the structure of solids and molecules.

3.2.1 ‘Third generation’ Synchrotrons

In the mid 1990s ‘third generation’ synchrotron sources became operational, a typical device being the European Synchrotron Radiation Facility[8] or the Argonne Lab. Advanced Photon Source[9], which uses positrons rather than electrons in its ring. These third generation devices are much brighter than earlier devices giving 10^{19} photons $\text{s}^{-1} \text{mm}^{-2} \text{mrad}^{-2}$ per 0.1% wavelength interval and operate between about 1 keV and 200 keV in x-ray energy.

3.2.2 Free Electron Lasers

A planned ‘fourth generation’ technology of accelerator based x-ray lasers is the x-ray free electron laser (x-ray FEL or sometimes denoted XFEL). This is a very different scale to the table-top sources but is interesting for comparison purposes. These devices promise to give powerful beams of coherent x-rays, probably exceeding the capabilities of table-top designs in performance but limited by being large scale facilities.

Free electron lasers (FEL’s) operate at a number (approximately 20) of facilities around the world. They typically operate in the millimetre wave to visible region of the spectrum. Their output can have the optical properties characteristic of lasers and produce high power beams. In a FEL the laser ‘medium’ is free electrons. These are moving at relativistic speeds along a beam line, they then pass through a periodic magnet (wiggler) section see Figure 1. Normally, this section is surrounded by a buildup cavity but for some configurations the buildup is done in a single pass. The magnetic field of the wiggler modulates the electron bunch transversely and if the phase matching is right, the electron bunch contributes some of its power to the co-propagating radiation field, hence providing gain. For a given wiggler structure, the wavelength is determined only by the energy of the electron beam. The Doppler shifts involved mean that the frequency of the emitted light is much higher than the modulation frequency of the electrons.

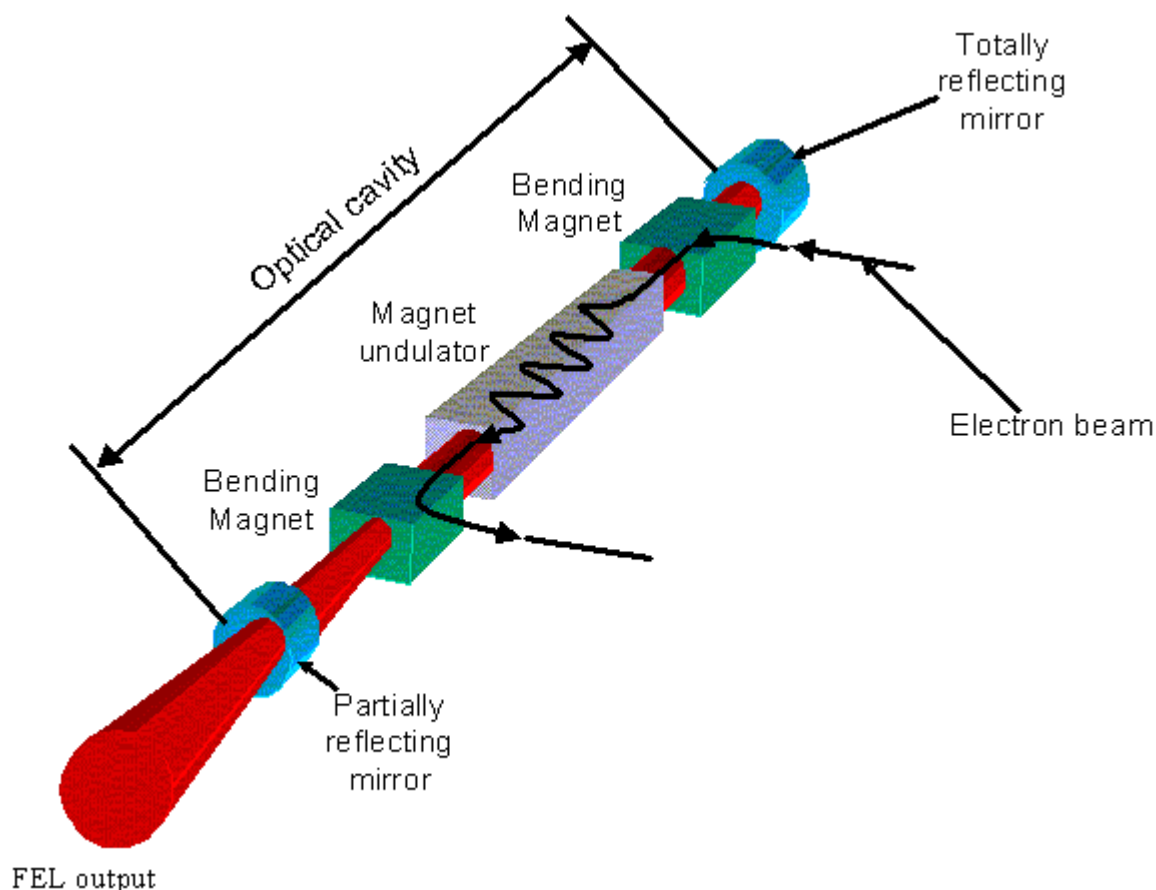


Figure 1 Schematic of operation of a free electron laser. Shown with a buildup cavity which is not used at high frequency.

At a number of accelerator facilities there are plans for x-ray FELs and a number of preliminary studies have been done at lower wavelengths to test the technology and the computer simulation codes.

Progress on these sources is rapid and ongoing. For example at SLAC in California a gain of 10^5 at $1.2\mu\text{m}$ was realised as a test for the linac coherent light source (LCLS) The LCLS is a multi-institutional proposal for a single-pass x-ray FEL operating in the 0.1-1.5 nm wavelength region, using electron beams from the SLAC linac at energies up to 15 GeV[10].

Similarly a VUV FEL for wavelengths down to 6 nm in the vacuum-ultraviolet and soft X-ray regime is under construction at the TESLA Test Facility (TTF) at DESY in Hamburg, Germany (TESLA is a proposed collider)[11]. At present, lasing has been observed down to 80 nm, the shortest wavelength ever achieved with a free electron laser. When fully operational this facility is expected to reach to a 6.4 nm in wavelength and have a brilliance of 10^{23} photons $\text{s}^{-1} \text{mm}^{-2} \text{mrad}^{-2}$ per 0.1% wavelength interval an increase of 10^{10} in the number of photons per pulse. In addition, an X-ray FEL laboratory for wavelengths just below 0.1 nm is proposed to be built within the TESLA project[12]. An x-ray laser initially covering 100 nm to 0.3 nm is proposed at MIT/Bates[13].

3.3 X-ray lasers

The quest for practical x-ray lasers began in the 1960's. However the dramatic scaling of pump power requirements along with the complexity of excitation schemes and low reflectivity of bulk materials which means that no multipass amplification is possible meant that it was 1984 before the demonstration of large amplification at soft x-ray wavelengths (for example [14]). Collisional excitation was first observed in neon-like selenium since then many other neon-like materials eg copper ($Z = 29$) to silver ($Z = 47$), have been made to lase [15,16].

There followed numerous demonstrations of x-ray lasers typically using outer shell transitions in highly ionised plasmas, but these required the most powerful optical lasers as pump sources. The early history and principles are covered in the book of Eltham[17]. X ray lasers have been the subject of a series of international conferences, the most recent being the 8th held in 2002[18]. A vast amount of operational detail is described in this set of proceedings.

Some specific new approaches to x-ray lasers are discussed in detail in a later section (4.1.3).

4 New Technologies and techniques

The coherence of the radiation from standard coherent x-ray sources is poor and techniques such as perturbing and filtering have to be used to and reduce the effects of incoherence such as poor contrast of images[19]. This is done at the cost of brightness. However, the new techniques for x-ray production can produce significantly more monochromatic radiation, greatly increasing its usefulness.

This section discusses new technologies for x-ray production and control. The Kerr-effect mode-locked laser combined with capillary arrays for confinement of a laser or harmonic generation, medium has led to improved harmonic generation, faster pulse generation and simplified x-ray lasers. Later in the section techniques of interaction with nuclear energy levels are discussed. The final subsection discusses the recent demonstration of an x-ray Fabry-Pérot interferometer.

4.1 Capillary technology

In lasers producing wavelengths shorter than about 50 nm the laser medium must necessarily be a plasma because of the high energy density requirement dictated by the use of atomic transitions. Plasma generation and the interactions of relativistic electrons is complex and detailed computer models have been developed to predict the behaviour of the plasma. In the first generation of experiments, powerful visible or infrared lasers produce the plasma from a solid target. In the last decade a new technique capillary technology where the plasma is confined in an array of capillary tubes, and using a pre-pulse before the main pulse to improve the pump laser to plasma coupling [20] have produced a table-top versions of these x-ray laser techniques. Capillary techniques have come to be used in a number of applications described in the following subsections.

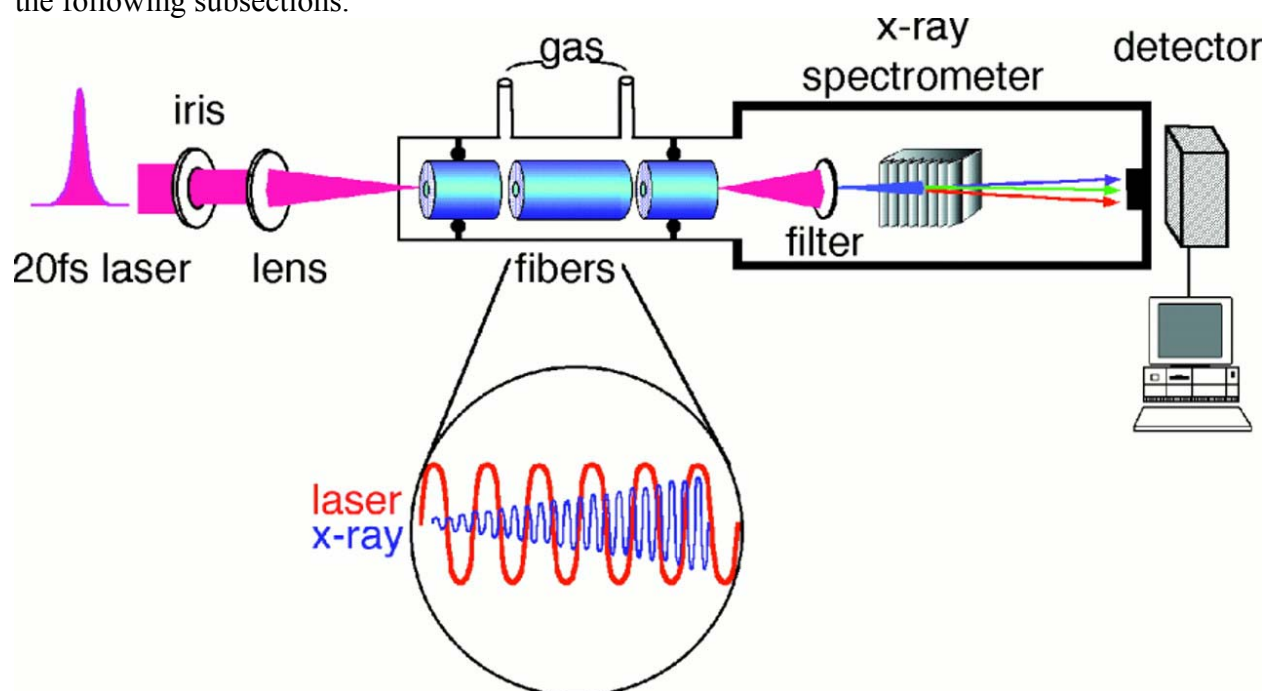


Figure 2 Apparatus for high harmonic generation in a capillary bundle. Inset shows the x ray buildup in the plasma. Note that the wavelengths are not to scale. From [21].

‘The field is now approaching the stage at which soft X-ray lasers sufficiently compact to fit on a normal optical table will be routinely utilised in science and technology’ [6].

4.1.1 High harmonic generation

For many applications a possible alternative to the very large expensive x-ray sources such as synchrotrons is the production of x-rays by harmonic generation.

By making use of the highly non-linear properties of materials very high harmonic generation can be achieved. The usual targets are gas, as the medium must be transparent to x-rays, or plasma generated at the surface of a solid; very high harmonics can be produced [21] but the methods of producing EUV light has until recently been very inefficient.

The development of Kerr-lens mode locking (also called self-mode locking) enabled the production of femtosecond pulses with high peak powers[22]. It is based on the Kerr-effect which is the non-linear intensity dependent index of refraction and has been demonstrated in various solid-state lasers and proved to be an effective and stable method for generating ultrashort laser pulses. In Kerr-lens mode locking the intracavity self-focusing effect is utilized to produce a bias on the transverse modes of the laser, such that the laser has higher gain for the pulse state than for the cw state. Because the optical Kerr effect is not sensitive to wavelength, the Kerr-lens mode-locking mechanism does not restrict the wavelength of the laser. For a detailed description, see Chapters 9 and 10 of Lasers, by Siegman[23].

From visible to 200 nm and perhaps beyond nonlinear optical crystals are the technology of choice for harmonic generation, but at higher energies these are no longer transparent or are damaged so the non-linear medium is by necessity a gas. The input is typically a pulse from a Ti:Sapphire laser that is of sufficiently high energy and short length to produce an electric field that can strip electrons from an atom. These electrons recombine when the electric field reverses causing the release of high energy photons. Two practical problems occur, phase matching the input and output light, and the spread of different harmonics produced. The phase matching condition has been overcome by the use of capillary arrays to contain the gas. This breakthrough was to use the change in phase velocity of light with wavelength in a capillary to match the phase [21] giving an increase of power of three orders of magnitude over the unmatched process see Figure 2. The second problem has also to some extent been successfully addressed. Shaping of the input pulse using the coherent control techniques described in section 4.2 allows the optimisation of the distribution of energy among the harmonics[24] as shown in Figure 3. That is the feedback in the optimisation routine is working towards maximising the power in a chosen harmonic. Using this method, they have enhanced the energy content of a specific mode by more than an order of magnitude.

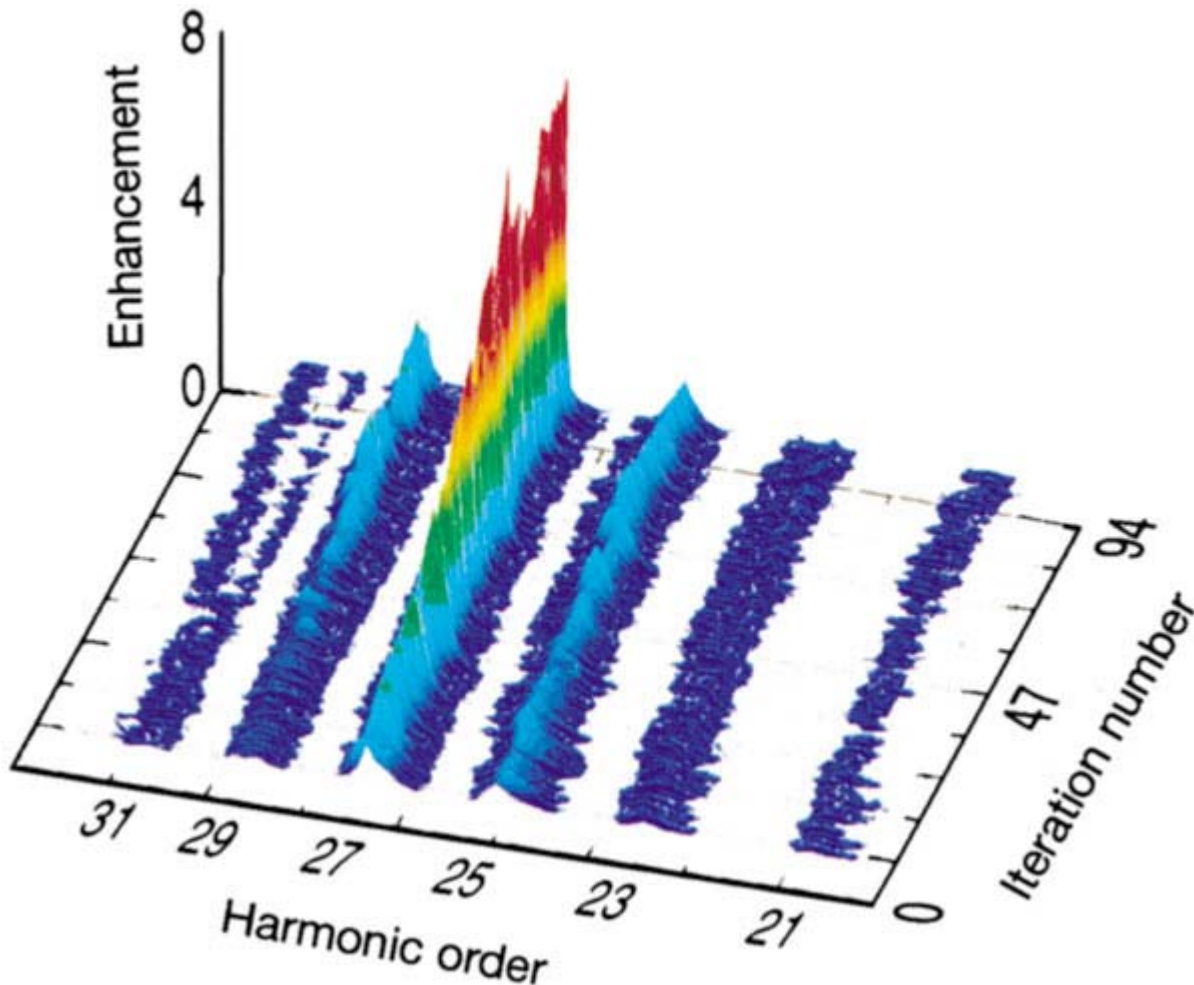


Figure 3 Showing the optimisation of the transfer of energy to the 27th harmonic in a frequency multiplication system[24].

Using a modulated capillary, that is modulating the core diameter of the fibre by 5-10% the same group of Kapteyn have produce high efficiency generation of EUV (50 eV, 25 nm)[25].

Recent cascaded schemes have improved the efficiency and range of high harmonic generation down to 160 nm[26].

4.1.2 Attosecond pulses

To generate exceptionally short pulses, in order for there to be a few cycles of the radiation in the pulse envelope then the carrier frequency must be extremely high. This puts a lower limit on the length of an optical pulse of the order of a few femtoseconds. The fastest pulses yet generated are in the attosecond regime and hence must be x-ray in wavelength.

A collaboration led by the group of Krausz at the Vienna Institute of Technology has reported the generation and detection of pulses in the attosecond regime[27]. Included in the collaboration is Drescher at Bielefeld University who has reported[28] a measurement the recovery from the ejection of an inner shell electron in a krypton atom. The experiment was done by varying the delay between an x-ray pump and an optical probe pulse. This is done using a variable path-difference to a claimed resolution of 100 as.

The pulses were generated by the action of femtosecond optical pulses on neon gas in a jet. High harmonic generation is a result of the electron-ion interaction following the strong field ionisation[29].

The x-ray pulse length produced is estimated to be 650 as.

This work has generated much interest; a reviewer in *Nature* saying, “We are entering a new realm of hyper-fast measurement – the age of attophysics has begun”.

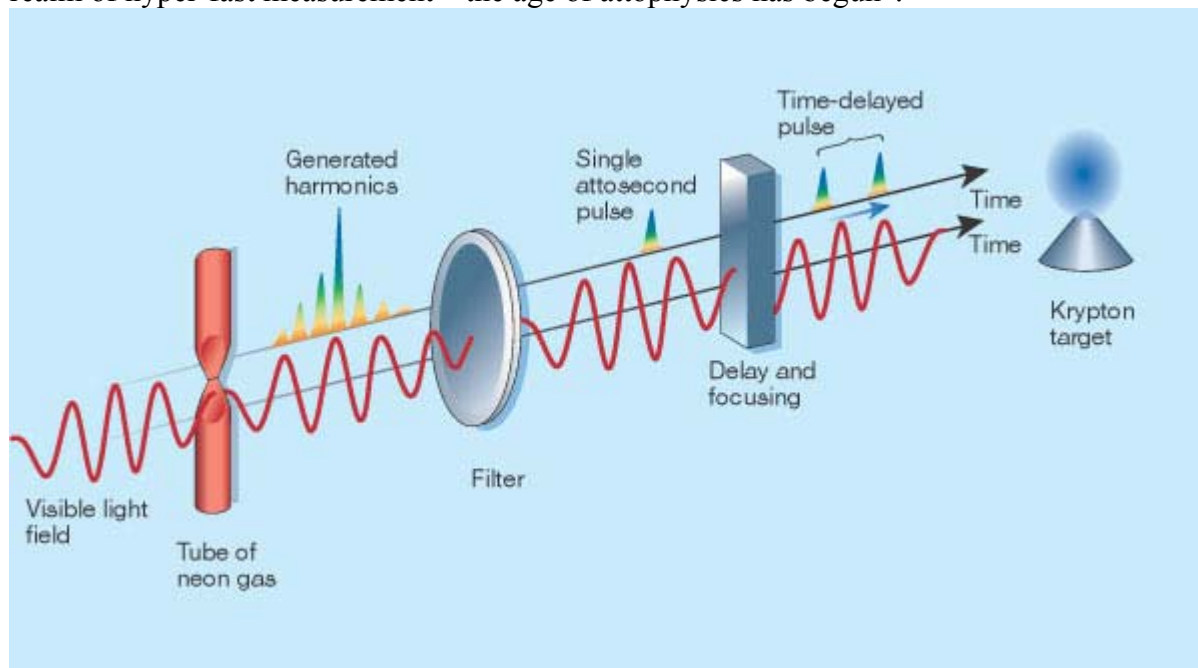


Figure 4 Generating and using attosecond pulses. In the work of Hentschel et al[27], harmonics of an ultrashort pulse are generated in neon gas and filtered to produce what is thought to be a single pulse, which is then used to monitor the generation of photoelectrons in krypton

There is some dispute over the strength of the evidence that a single pulse has been produced. However, these and other workers have clearly demonstrated trains of attosecond pulses. What the Krausz group has shown is time resolution in the attosecond regime.

The work will in itself improve the development of x-ray laser techniques by increasing the knowledge of the detailed behaviour of inner shell electrons in high fields.

It is seen that the x-ray pulse is locked to the carrier wave of its generating optical pulse. The authors of [28] attribute this to either the high-harmonic generation process suppressing the phase variation from pulse to pulse or, an option they consider more likely, is that only when the phase is optimal is an x-ray pulse generated. In this case stabilising the phase of the driver pulses would dramatically increase the yield of x-rays.

This has been followed by a very recent paper[30] where control of the carrier phase of light has been demonstrated using feedback stabilisation of phase shifts in the amplifier stages. The pulse train from a mode locked laser amplified in this manner is used to produce soft x-rays in a jet of neon. They have confirmed predictions that the highest energy photons show a critical sensitivity to the carrier-envelope phase, this is because the highest energy interaction can only occur if the electric field evolves in the right form during the atom-field interaction. Turning off the phase control washed out the features in the spectrum. Using this technique, the authors claim that they

can trigger and steer the motion of an electron with a precision limited only by quantum mechanical uncertainty.

These techniques can be used to make measurements of extremely fast processes that have not previously been accessible.

An earlier paper[31] gives more experimental details on the phase delay technique (but still not much). The source described here is a Kerr lens mode lock Ti:sapphire laser giving 9 fs 20 nJ pulses at 24 MHz. These pulses are broadened in standard single-mode fibre and doubled in BBO then compressed in a chirped mirror compressor giving 6fs 6nJ pulses. This paper also emphasises the links with the femtosecond comb, the phase control being needed either to produce the time resolved attosecond pulses or to lock up the comb.

A collaboration co-ordinated by Imperial College has been given a Research Councils' Basic Technology Programme grant to develop an attosecond facility[32].

4.1.3 X-ray capillary lasers

Rocca *et al* working at Colorado State University revolutionised the x-ray laser field in the mid 1990's with the demonstration of a table-top soft x-ray laser using capillary discharge[33,34]. This source is compact and practical compared to the giant devices that preceded it. This laser uses a fast discharge excitation of a gas-filled capillary channel. It emits at 46.9 nm with an average power of several milliwatt. The spatially coherent average power per unit bandwidth generated by this laser is comparable to that emitted at this wavelength by undulators at third generation synchrotron facilities, and its coherent peak power is six orders of magnitude larger. Since the initial development sources have been developed using alternative laser lines. The development of these lasers has now reached a point at which these sources can be employed in new exciting scientific applications ranging from materials science to the diagnostics of dense plasmas.

Two principle new technologies have enabled the development of table-top soft x-ray lasers. The first is pumping of x-ray lasers by high power ultrafast optical lasers using chirped-pulse amplification. These lasers are typically based on Ti:sapphire, Nd:glass or Cr:LiSAF. The chirped-pulse amplification technique involves stretching a seed pulse in length then amplifying and recompressing to near the original length thus avoiding high peak power in the amplification system[35]. The discovery of Kerr-lens mode locking and its subsequent refinements have made the generation of sub 20fs seed pulses routine.

The second important new technology is the introduction of the capillary discharge as is used in high harmonic generation methods described in section 4.1.1. Instabilities in the gain plasma have long been a problem in x-ray laser development. This has changed with the introduction of capillary discharges where the plasma is contained in channels 1.5 mm - 4 mm in diameter, filled with preionised material.

The reference by Backus *et al*[35] reviews lasers based on chirped pulse amplification and Rocca[6] reviews the status of soft x-ray lasers based on population inversion in plasmas.

4.2 Pulse shaping – coherent control

A number of techniques described in this section rely on the ability of produce pulses of the required shape. Techniques for producing tailored pulses include using a liquid crystal modulator acousto-optic modulation and using deformable mirrors (for a review of methods see Weiner[36])

The pulse shape is manipulated by dispersing the pulse with a grating and then using a phase modulator or deformable mirror in the frequency domain, then recombining the phase shifted components on a second grating to produce a modified pulse.

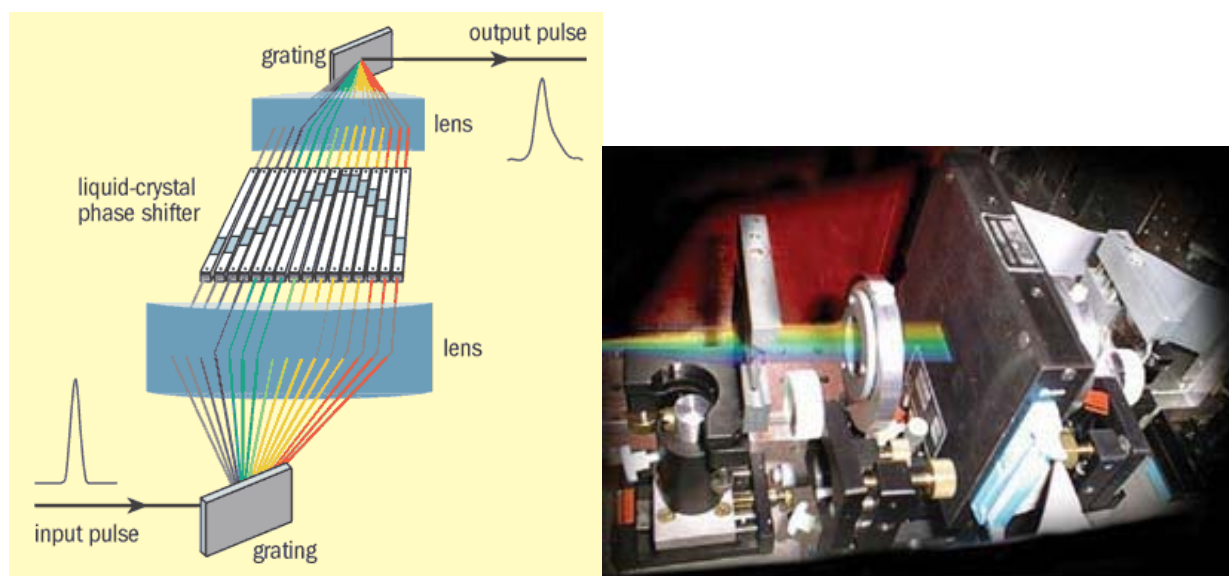


Figure 5 Schematic and photograph of a coherent control system based on a liquid crystal phase shifter developed by the group of Gustav Gerber at the University of Würzburg, Germany. From Opto & Laser Europe March 2003[38].

Chemical physics studies are benefiting from the ability to produce tailored pulses in the deep UV corresponding to a range of molecular absorptions. This allows the control coherent of molecular reactions[37], that is control of chemical synthesis in a bond-selective way by controlling the shape of the wavefunction. This ability may also have applications in quantum computing.

In the coherent control method an initial guess is made as to the shape of the optical pulse that will produce the required output. For example excite the reactant molecules in such a way as produce the desired reaction products in the case of tailored chemical reactions or produce the required harmonic in the case of high harmonic generation. The pulse is generated and the result analysed. Usually by mass spectrometry for a chemical reaction and by x-ray spectroscopy for harmonic generation. This data is used to refine the pulse shape by a learning algorithm using one of the modulation methods described above.

A related technique is the coherent modulation of x-ray pulses. This was demonstrated by the group of Reis[39]. Their method can switch the picosecond pulses produced by synchrotron sources and they hope to produce sub-picosecond switching. The method is based on the Borrmann effect, where the usual strong attenuation of x-rays in solids is dramatically reduced in a crystal if the lattice planes are oriented in such a way as to act as a waveguide. The Borrmann

effect is very sensitive to lattice variations so an acoustic wave in the crystal, generated by a femtosecond pulse, temporally destroys the effect along the wavefront of the acoustic pulse. This technology is close to producing an x-ray version of the acousto-optic modulator which is used in optical locking techniques. Thus x-ray sidebands may be produced which would be a major step forward in coherent x-ray frequency metrology and necessary for example to allow locking of an x-ray source to a nuclear transition.

4.3 Mössbauer effect frequency references

Radiation from nuclear transitions has very narrow natural linewidth. The Mössbauer effect is the process whereby nuclei of atoms imbedded in solids undergo recoilless transitions, and hence can emit and absorb low energy gamma rays of the natural linewidth. This is a process that is naturally Doppler free, in a similar way to an ion in a trap when confined in the Lamb-Dicke regime. The energy of gamma rays is such that the momentum of one photon is significant on the scale of the natural linewidth and Doppler broadening. In a recoilless transition the atom is bound in the crystal and the momentum is taken up by the crystal as a whole. The energy of recoil is shared between the gamma ray and the phonons and a Mössbauer transition occurs if the state of the lattice remains unchanged.

The classic example of a Mössbauer transition is that from the first excited state of the iron-57 nucleus which is at an energy of 14.4 keV (wavelength of 0.08 nm). It has a lifetime of 141 ns and hence a 4.8 neV width. This is a Q factor of 3×10^{12} far in excess of atomic resonances, other Mössbauer lines have Q values in excess of 10^{20} . The frequency of the Mössbauer resonance is very stable, the stability is of the order of 10^{11} without any special precautions, because it is a nuclear process and the nucleus is naturally well isolated inside the atom. Only the weak hyperfine coupling links the external influences acting on the electrons to the nucleus. Compare this with the silicon lattice constant, which is known to 2.9 parts in 10^8 but requires very precise control of temperature (~ 1 mK stability) and pressure to reproduce. Reference x-ray wavelengths that have been measured using x-ray diffraction are known to only a few parts in 10^7 .

It has been proposed that the Mössbauer effect could be coherently controlled[40]. This could lead to a new branch of spectroscopy, laser-Mössbauer spectroscopy, and would be a possible method for the realization of a gamma-ray laser (see section 4.4).

In recent work using a silicon crystal reference, the iron-57 Mössbauer line was determined to 1.9 parts in 10^7 [41], so the absolute accuracy is much less well known than the stability would allow. The experiment was done by resonantly exciting the Mössbauer transition using a synchrotron x-ray source and comparing the wavelength of the re-emitted radiation from the Mössbauer source with backscattered radiation from silicon crystal planes, which have wavelengths known in terms of the silicon lattice constant. This experiment was enabled by brighter sources of x-rays from synchrotrons and the prospect of x-ray laser sources with lower beam divergence gives the expectation of another big advance in resolution.

This measurement was limited by its use of a silicon crystal as a length reference. However a combined x-ray and optical phase interferometer could provide a much more accurate x-ray length standard (see section 4.6). This is important in applications such as nanometrology where lengths small compared to a wavelength of light must be measured. Here well characterised radiation is needed, not an etalon of well defined material.

A link between fundamental constants and x-ray frequencies (or wavelengths with equal accuracy as the speed of light is exact) may be provided by hydrogenic systems. These are atoms or ions with a single electron and hence have a calculable electron structure. The classic example is hydrogen where the Rydberg constant provides a natural frequency scale linked to atomic constants. In more exotic systems, such as He^+ and higher atomic number highly charged ions, some of the transitions that are in the optical region of the spectrum of hydrogen move into the EUV or x-ray. These could be used as calculable frequency standards allowing measurements made at optical together with theoretical knowledge of the atomic frequency interval to give precise x-ray frequency references.

4.4 Gamma-ray lasers: ‘Grasers’

Over the years there have been many proposals for ultrashort (<1 nm) wavelength lasers based on various schemes mainly using the Mössbauer effect. These proposed devices have been known as gamma-ray lasers or grasers. There has also, more recently, been experimental work towards realising this goal, often funded by defence applications. The complex issues of pumping mechanisms are reviewed by Baldwin and Solem[42].

Although much more difficult to realise than an optical laser the gamma-ray laser would in theory have a higher power and much smaller divergence and hence a higher brightness by many orders of magnitude than could be obtained in other lasers.

The use of nuclear transitions has some advantages, excited states of nuclei have much higher energies than those of atoms and hence the pump power needed to generate a similar energy density is reduced. In most graser proposals a solid state laser material is pumped by nuclear reactions. The Mössbauer effect enhances the interaction of radiation with resonant nuclei and the Borrmann effect reduces the losses that normally suppress the gain. However there are many practical difficulties, such as the sensitivity of the Mössbauer and Borrmann effects to temperature and the competing non-radiative decays in many systems. In summary, although some approaches show promise, the goal of a 1 nm laser source is still far away, at least a decade behind other x-ray lasers[17].

4.5 Nuclear isomer excitation

In a similar way to electron excitation in atoms, nuclei can be excited to higher internal energy states; the transitions between these states produce gamma rays. Some of these nuclear states have unusually long lifetimes, and these are known as nuclear isomers. These have been studied as they hold the promise of energy storage and of gain for a gamma-ray laser, but even if the problems associated with producing gain meant that the goal of a gamma-ray laser was not achieved, there is still the possibility of using these levels as ultra-narrow x-ray frequency standards.

The principle long lived isomers studied are hafnium-178 (energy 2.4 MeV, half-life 31 years)[43], tantalum-180 (energy 75 keV, half-life 1.2×10^{15} years)[44] and thorium-229 (energy 3.5 ± 0.1 eV, half-life 45 hours)[45] an exceptionally low energy for a nuclear transition corresponding to 350 nm. If laser radiation could be used to stimulate this transition then a system of frequency references potentially as stable as ion traps could be envisaged (as well as a gamma ray laser which motivates the investigators). The photo-induced depopulation of an isomer of tantalum-180 has been made[46] and confirmed [44]. It was thought that the decay rate of the hafnium-178 transition had been increased by irradiation [47] but it now seems that this observation may have been premature [43].

The use of nuclear isomers as a power storage system has been suggested and is being considered for military applications[48].

This field is sometimes known as ‘quantum nucleonics’ or ‘opto-nucleonics’ a recent review of experimental results and concepts has been published by Carroll *et al*[49]

4.6 X-ray phase interferometry

An x-ray Fabry-Pérot interferometer has been demonstrated for the first time in 2003[50]. This may lead to a range of new x-ray devices using phase interference. Potential applications include calibrated nanoscale length metrology, x-ray clock, x-ray interference filters and perhaps even locking of x-ray sources to cavities as is done with x-ray lasers or locking of cavities to x-ray sources that are naturally very high Q such as Mössbauer resonances.

A Fabry-Pérot consists simply of a pair of parallel reflecting surfaces. The device demonstrated consisted of parallel sapphire surfaces. A single crystal sapphire can produce Bragg backscattering of x-rays in a similar way to a multilayer mirror in optics. This is in contrast to conventional x-ray mirrors where only grazing incidence x-rays are reflected. Earlier work[51] used silicon crystals which proved to have lower reflectivity, but still showed storage of energy. The disadvantage of the Bragg reflection method is that, like optical multilayer mirrors, the x-ray mirrors only operate over a narrow spectral range. The interferometer demonstrated used x-rays from a synchrotron source at 14.3 eV chosen to match an available Bragg reflection in sapphire but also to be close to the iron-57 Mössbauer resonance. A finesse of 15 and 0.76 μeV wide fringes were observed, this is a spectral resolution 100 times better than a crystal monochromator. This result is only a considered a demonstration of the principle rather than a working tool; overcoming the difficulties of preparing faultless crystals and aligning them with the parallelism needed ($\sim\mu\text{rad}$) is very impressive and the experiment clearly shows the potential of an x-ray interferometer. A combined x-ray and optical Fabry-Pérot could directly measure x-ray wavelengths in terms of optical ones, rather than using the silicon lattice constant as is done at present and so offer enormous potential improvement in x-ray wavelength metrology. A second application of the interferometer would be as a narrow filter for dynamic versions of the x-ray studies of the types discussed in (2.2).

5 Summary: Advantages and potential of coherent x-ray sources

Coherent x-ray sources will increase the utility of x-rays both in areas of current use and by opening up new applications.

A new type of source is being developed. These are constructed on laboratory sized optical tables and given the name 'table-top' to distinguish them from accelerator based x-ray sources that are facility sized. The table-top sources have come about as a result of the development of Kerr-effect mode-locked lasers as a source of high power femtosecond pulses, combined with capillary arrays for confinement of a laser or harmonic generation medium. This has led to improved harmonic generation, faster pulse generation and simplified x-ray lasers. Along with advances in soft x-ray optics, these sources will open up a range of applications.

Accurate frequency metrology for x-ray spectroscopy will become possible, dramatically improving calibrated length measurement at the atomic scale.

Further into the future, perhaps over a ten year timescale, there will be the development of x-ray phase interferometry and other techniques usually associated with the optical regime such as sideband locking. Even further ahead this can be expected to lead to x-ray clocks, perhaps based on calculable transitions in atomic systems, or nuclear transitions. In these systems, locking an x-ray laser to a Mössbauer transition would give the same advantages over optical transitions and an equal or greater magnitude of resolution improvement, that optical transitions have produced over radiofrequency ones

A technologically parallel area is the production of attosecond timescale pulses, for which the carrier is by necessity an x-ray frequency. Attosecond pulses will initially be used for studies of ultrafast processes in atoms but will have metrological applications particularly in synchronisation and in very high time resolution measurements.

Dramatically increased brilliance of x-ray sources will allow structure determination from smaller and non-crystalline samples of materials, which would have wide industrial applications and be of particular interest in chemical analysis for biotechnology.

New, very large scale, light sources are being designed and built in the UK as well as in Europe, Japan and the USA. In the applications described above table-top systems will have applications where the use of a large scale facility is impractical or prohibitively expensive, and metrological and time and frequency applications are likely to come into that category.

Appendix

X-ray wavelength designations

This is a guide to the canonical designations of x-ray wavelengths but it is necessary to be aware that these are not universal and some authors use different values for the boundaries.

	Wavelength range /nm	Equivalent in energy units
Vacuum ultraviolet	200-100	6.2 eV-12.4 eV
extreme ultraviolet EUV,XUV	100-10	12.4 eV-124 eV
start of x ray	~ 30	~40 eV
soft x-ray	30-0.2	41 eV-6.2 keV
soft x-ray laser coverage	49-5.6	25 eV-221 eV
hard x ray	0.2-0.01	6.2 keV -120 keV

The SI unit for wavelength is the nanometre but still in common use is the ångström (symbol Å, 1 Å=10⁻¹⁰ m). For high energy radiation the photon energy in electronvolts is often quoted (1 Hz/h=4.135×10⁻¹⁵ eV)

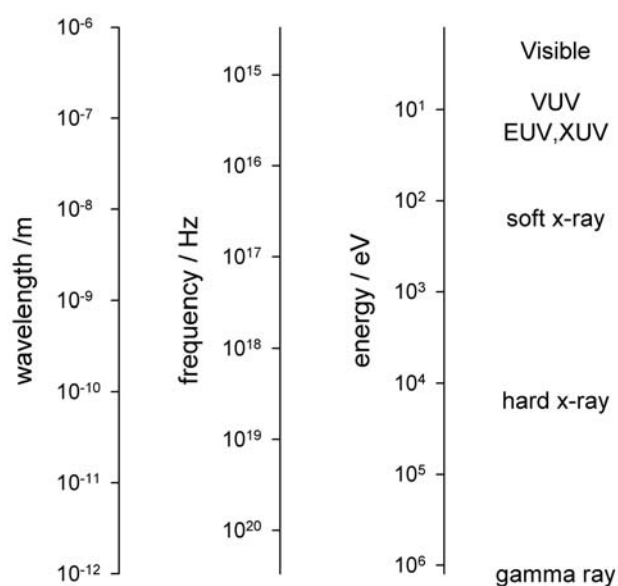


Figure 6 Relationship of the x-ray region band names to their values in different units

Glossary

The 'water window' is the range between the K-absorption edges of oxygen (2.3 nm) and carbon (4.4 nm). In this window x-rays highlight the difference between organic matter and water and so it is especially useful for investigating organic specimens

The Borrmann effect is a classical diffraction effect that allows the transmission of x-ray energy through very thick crystals (i.e. many times the incoherent x-ray absorption depth)[52].

The Mössbauer effect is the process whereby nuclei of atoms imbedded in solids undergo recoilless transitions and hence can emit and absorb low energy gamma rays of the natural linewidth, that is with no Doppler broadening.

Flux is the number of photons per second passing through a defined area, and is the appropriate measure for experiments that use an entire, un-focused x-ray beam.

Brilliance of a source is one of the most important figures of merit. The brilliance is a measure of the intensity and directionality of an x-ray beam, measured in number of photons per second in a narrow energy band-width and in a unit of solid angle typically quoted in $\text{photons s}^{-1} \text{mm}^{-2} \text{mrad}^{-2}$ per 0.1% wavelength interval. It determines the smallest spot onto which an x-ray beam can be focused.

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