Laser-produced X-ray sources

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1. Introduction

Since the discovery of X-rays by Roentgen in 1895, scientists have been producing laboratory X-rays by accelerating (and decelerating) electrons in evacuated tubes. In recent years, however, a disparate variety of laser methods have been developed that produce laboratory X-rays with new characteristics, including spatial and/or temporal coherence, variable time structure and energy, and very-high-field or high-energy densities. In some of these light–matter interactions, extreme physical regimes are being realized for the first time in laboratory settings, guaranteeing a rich variety of forthcoming physical insights and applications. At first glance it seems remarkable that 1.5 eV laser photons can be used to produce X-rays of order 1 keV (high-harmonic generation; X-ray lasers), tens of keV (inverse-Compton sources; MJ-class laser-produced plasmas), MeV (PW-class laser-produced plasmas), and even GeV (laser-wake techniques). In this overview, we briefly describe these six methods of producing X-rays with modern laser techniques (tabulated below), pointing out how they provide radiation with novel characteristics that add value in both fundamental and applied investigations. This is not intended to duplicate the excellent review papers that have already been produced in some of these areas (Pfeifer et al., 2006; Brabec and Krausz, 2000; Eden, 2004; Umstadter, 2003; etc.), but rather to survey on behalf of the larger radiation physics community (potential consumers) these new tools, techniques, and mechanisms of producing X-rays that will surely only gain in prominence going forward. Finally it is stressed that metrological care, usually in the form of calibrated diagnostics, is a necessary contributor to the success of these investigations, and ultimately, the medical, alternative-energy, and device applications that will emerge.

2. Sources of laser-produced X-rays

2.1. Megajoule-class laser-produced plasmas

The study of extreme states of matter is motivated by the search for alternative-energy sources, hard X-ray imaging applications, nuclear weapons effects and testing, and basic physics research, including atomic, plasma, hydrodynamics, astrophysical, and materials phenomena. In the next decade, one of the primary goals in high-energy-density (HED) physics is to achieve controlled thermonuclear ignition. Many facilities around the world are working to support this effort, and two facilities that expect to achieve ignition are le Laser Mégajoule (LMJ) of the Commissariat à l’Energie Atomique (CEA), being constructed near Bordeaux, France and the National Ignition Facility (NIF) under construction at the Lawrence Livermore National Laboratory.
The strategy of laser-driven inertial confinement fusion (ICF) is to focus ultraviolet lasers onto capsules filled with heavy hydrogen. In indirect-drive ICF, the capsule is suspended within a metallic cylinder called a hohlraum. The hohlraum is heated to hundreds of eV, creating a high-Z plasma with an electron temperature of several keV. X-rays emitted from this plasma ablate the capsule’s exterior polymer shell, compressing the fuel at velocities up to 300 km/s and to particle densities of 10^{25}/cm^3. The baseline hohlraums and capsules of NIF are expected to require at least 1.35 MJ of laser energy to achieve ignition. This would be delivered in 192 beams at over 400 TW, exceeding the output of the global power grid for a few nanoseconds. At ignition the core could reach \( >30 \, \text{keV} \) (Lindl et al., 2004), producing many high-energy electrons, X-rays, neutrons, as well as electromagnetic interference and debris. Clearly, this is a physically hostile environment providing challenges to the array of needed diagnostic instrumentation (Hudson et al., 2006).

### 2.2. Petawatt class of laser-produced plasmas

Today, solid-state lasers can attain peak pulse powers of terawatts with table-top systems, and up to petawatts at larger-scaled facilities. Among the most popular use today are solid-state lasers that employ Ti-doped sapphire (Ti:Al_2O_3, \( \lambda \approx 806 \, \text{nm} \)) as the gain medium. They possess a very broad gain bandwidth, that is, they support a large number of discrete longitudinal modes or frequencies in the lasing cavity. Since output-pulse duration is inversely proportional to the number of modes that are phase locked, it is then possible to produce ultrafast, few-cycle pulses possessing up to a joule of energy (Table 1).

Two key developments have permitted the delivery of ever-more intense laser output into increasingly smaller domains of space and time, which in turn has permitted nonlinear and relativistic interactions with matter leading to X-ray production. Indeed, the techniques discussed in the remainder of this paper are all rooted in these landmark breakthroughs. The first is chirped pulse amplification (CPA, Strickland and Mourou, 1985), a technique wherein short pulses from a laser oscillator with durations ranging from a few fs to ps are dispersively stretched in duration (spatial chirp), amplified, and then recompressed to produce intensities ranging from \( 10^7 \) to \( 10^{21} \, \text{W/cm}^2 \). The second key development is the use of pulse shaping with adaptive optics or various dispersion-compensation techniques that permit focusing of a laser beam to approach the diffraction limit. Using such laser pulses to produce X-rays, powerful probes matched to both the spatial and temporal scales of nano-world dynamics can be produced. Indeed, these new laser-produced X-ray techniques provide the best of both worlds: the high energies of X-rays combined with the time structure, intensity, polarization, and coherence of lasers.

There are a wide variety of ways in which ultra-short, ultra-intense laser pulses can be used to create X-rays, usually through controlled acceleration and deceleration of electrons. Laser light interacting with a gas, for example, with relatively low field strengths, causes bound electrons to oscillate. This acquired “quiver energy” of an electron in a time-varying electric field is proportional to the field strength; the time-averaged energy is called the ponderomotive energy. At laser-field strengths around \( 10^{15} \, \text{W/cm}^2 \), ionization and acceleration of free electrons can then occur (Umstadter, 2003). In the case of interaction with a solid surface, laser light can initially produce and then interact with dense plasmas. Energy can be readily transferred from the laser field into the plasma through the acceleration of free electrons. When laser intensities produce electron quiver energies comparable to the rest mass energy of the electron, e.g. at \( 2 \times 10^{18} \, \text{W/cm}^2 \) at 800 nm, the resulting laser–plasma interactions are said to be relativistic. Such hot electrons can reach MeV energies, and propagate both into solid targets and into vacuum. The latter group strongly interacts with electromagnetic radiation in a process called vacuum heating (Berezhiani and Shاتashvili, 2006) and many of these electrons are drawn back toward the target due to a space charge field established by the depletion of electrons from the interaction spot. This in turn can produce very short X-ray pulses containing both hard characteristic X-rays and continuum. Hot electrons are also produced through reflexing electrons in irradiated solids, magnetic-Lorentz-force heating in relativistic plasmas, and through inverse bremsstrahlung and resonance absorption in both relativistic and non-relativistic regimes (Wilks and Krueer, 1997).

One high-profile application that involves both high-energy (MJ) and high-power (PW) lasers is the attempt to produce inertially confined fusion in the laboratory. One idea that is being actively pursued at various laboratories around the world is fast ignition (Tabak et al., 1994; Kodama et al., 2001; Town et al., 2005), where the compression of a heavy-hydrogen target is accomplished with many simultaneous long-pulse (several ns) laser beams and the heating required to initiate fusion is performed by short (\( \approx 10 \, \text{ps} \)) intense laser beams after the target has reached peak compression. The generation of MeV hot electrons (Wharton et al., 1998; Yasuike et al., 2001) due to ps laser–matter interactions also leads to the production of X-rays as they slow down in a material, producing line emission by inner-shell ionization and a hard-bremsstrahlung continuum. Hence, another common use in ICF research of short, intense laser beams is the irradiation of a secondary target, producing high-energy X-rays, typically \( >100 \, \text{keV} \), which can backlight the primary, driven target and produce snapshot radiographs of its evolution with good temporal resolution. However, the MeV electron

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**Table 1**

Laser-produced X-ray sources.

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<tr>
<th>Novel X-ray source</th>
<th>Some distinguishing characteristics of X-ray emissions</th>
<th>Some X-ray applications</th>
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<td>Pulses of ns duration, incoherent, high-energy-density (HED) science</td>
<td>Inertial-confinement fusion, nuclear weapons effects and testing</td>
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<td>PW-class laser produced plasmas</td>
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circulation outside the small (≈ 10 µm) laser focal spot can result in an unacceptably large source size, which can adversely affect the radiogram’s spatial resolution (Tommasini et al., 2008; Park et al., 2008) unless care is taken with target design.

2.3. X-ray lasers

As discussed above, optical lasers have been used in the exploding-foil technique to create heated plasmas, hot electrons, and highly charged ions. With carefully selected atomic systems and methods, collisional excitation of the pumped plasma can result in a transient population inversion (into excited states of the highly charged ions) and amplification of spontaneous emission, producing a soft X-ray laser (Rosen et al., 1985; Matthews et al., 1985). X-ray lasers have evolved from under-ground systems initiated by nuclear explosions to above-ground systems using high-energy laser drivers, to table-top high-peak-power laser systems that use multi-pulse schemes to divide the work of plasma creation, electron stripping, and excitation (Dunn et al., 1998). Recently it has been shown that high-harmonic laser pulses (discussed below in 2.6), tuned to a soft X-ray laser’s transition energy, can themselves be amplified in the collisionally excited plasma amplifier while preserving their desirable properties of high spatial coherence, low divergence, polarization, and short pulse widths (Wang et al., 2008). In addition, the narrow linewidth of the plasma amplifier increases the temporal coherence of the output, providing a bright tool for fully coherent probes and imaging below 20 nm (Wang et al., 2008).

X-ray free-electron laser (XFEL), based on the self-amplified stimulated emission of accelerated electrons being pursued at facilities such as the Free-electron LASer in Hamburg (FLASH) and the Linac Coherent Light Source (LCLS) at the SLAC National Accelerator Laboratory, is also worth mentioning. Here the lasing medium itself is the single-pass, accelerated beam of GeV electrons in an undulator system that interacts with its own synchrotron radiation, leading to microbunching and hence pulsed X-rays with variable time structures down to the femtosecond scale. These are expected to produce spatially coherent X-rays tunable up to several keV and with unprecedented laboratory-source brightness (10^12–10^15 photons/pulse in a 0.2%–0.4% energy bandpass). Planned applications include studies of plasma and warm-dense matter, structural studies on single biomolecules, femtochemistry, and studies of nanoscale dynamics in condensed matter (Shenoy and Stöhr, 2000). While not a laser-produced photon source, the temporal coherence of free-electron lasers (FELs) can be greatly enhanced by injection seeding with conventionally produced laser pulses; similarly, XFEL research is pursuing seeding with high-harmonically generated ultraviolet- and X-rays (see Section 2.6, and Lambert et al., 2008) or X-ray lasers that are tuned to resonance.

2.4. Laser-wake techniques

Radio-frequency linear accelerators have long been used for high-energy X-ray production. With the advent of short-pulse (≪ 1 ps) ultrahigh intensity (≥10^18 W/cm^2) lasers, a new method of electron acceleration to very-high energies has been developed called laser wakefield acceleration (LWFA). There are a variety of implementations and terminology in this rich, new field of laser–plasma interactions. A plasma wave (wakefield) can be created by a single pulse and driven by a train of pulses from such an intense laser propagating through an underdense plasma, that is when the laser wavelength λ_l is related to the intrinsic plasma frequency ω_p (ω_p^2 = 4πe^2n_0/m, where e, m, and n_0 are the charge, relativistic mass, and density of electrons in the plasma) as λ_l < λ_p = 2πc/ω_p, where c is the speed of light. The ponderomotive force due to the laser pulse expels electrons from a region in its wake, exciting a transverse (to the laser-pulse direction) plasma wave, whose phase velocity is about that of the laser pulse group velocity (Esarey et al., 1996). There are schemes in which the plasma wave can be efficiently coupled to a train of laser pulses by adjusting the interpulse spacing and/or pulse width, τ, to be resonant with (Umstadter et al., 1994) or shorter than (Pukhov and Meyer-Ter-Vehn, 2002) the plasma period, i.e. τ ≈ 2π/ω_p. If the plasma wave is of sufficient amplitude, analogous to water waves it can “break” and electrons that were oscillating transversely at ω_p can begin to travel longitudinally (surfing the plasma wave) behind the laser pulse and be continuously accelerated up to very-high energies. Schemes have also been devised to inject electrons into the wakefield (Faurel et al., 2006) and, there are regimes where the large electric (wake) field continuously traps and accelerates background electrons (Pukhov and Meyer-Ter-Vehn, 2002). One recent demonstration of LWFA employed a 100 TW class Ti:Al_2O_3 laser directing 27 fs pulses (7.3 J) onto a supersonic He gas jet, producing quasi-monochromatic, multi-hundred MeV electron beams with an acceleration gradient of 1.6 GeV/cm, more than three orders of magnitude larger than in conventional radio-frequency linear electron accelerators (Yoshitama et al., 2008). It has also been noted that such injected and accelerated electron bunches are phased with respect to the driving laser, hence the suitability of such a plasma-cathode electron gun for pump–probe experiments (Umstadter, 2003) involving the laser and the electrons or X-rays produced by the same.

At present, laser irradiances can approach 10^{23} W/cm^2 and, when interacting with plasmas, can produce ultrarelativistic electrons. In this regime, laser–wake accelerated electron bunches can take the form of a parabolic flying mirror that, it has been proposed (Bulanov et al., 2003), can in turn interact with a counter-propagating laser pulse to focus, compress, scatter, and frequency upshift the light (Einstein, 1905), producing a power density such that the field strengths may exceed the so-called Schwinger “limit”, where vacuum begins to boil into electron–positron pairs (Schwinger, 1951). The first measurements of X-ray creation using relativistic paraboloidal shells of laser-accelerated electrons have produced a frequency multiplication of more than 100 (Kando et al., 2007).

Finally, in a related mechanism, relativistic electrons can be generated not only through interaction with a laser wakefield but also directly with the laser field itself via the inverse FEL mechanism (Pukhov et al., 1999). This method of accelerating electrons and potentially making hard X-rays is operative in the regimes of higher-plasma densities and longer–laser pulses than those where LWFA is operative.

2.5. Inverse-Compton sources

When a photon interacts with an electron whose kinetic energy is large compared to the photon energy, it may scatter from the electron and gain energy at the expense of the electron. This has been called linear inverse-Compton scattering and has been the idea behind a novel source of tunable X-rays produced by crossing laser and electron beams (Huang and Ruth, 1998). Such sources of X-rays have been demonstrated and the formalism developed to describe various aspects of performance has been verified quantitatively (Brown et al., 2004). The appeal of such sources derives from the ability to obtain tunable, short-pulse X-rays from a relatively compact system compared to the use of a synchrotron facility. This semi-monochromatic (a few percent bandwidth) source of X-rays is initially finding its greatest interest in medical-imaging applications.
For the case of a head-on collision of laser photons and relativistic (tens of MeV) electrons, X-rays are emitted in the direction of the electron beam, forwarded focused into a few milliradians. The X-rays are energetically dispersed about this forward direction in cones with half angle \( \theta \), given by \( E_\theta = E_0 \chi^2 / (1 + \chi^2 r_0^2) \), where \( E_0 \) is the laser photon energy and \( \chi \) is the relativistic factor for the electrons of energy \( E_0 \), viz., \( \chi = 1 + E_0[\text{MeV}] / 0.511 \). In this backscattering geometry, the X-ray burst is usually determined by the length of the electron bunch. By crossing the electron and laser beams at 90°, 300 fs X-ray pulses have been observed at 31 keV (Schoeplein et al., 1996, and called relativistic Thomson scattering). While this geometry sacrifices a factor of two in peak X-ray energy, the time structure offers a tool for fast pump–probe studies, including for example, the structural dynamics of materials.

Finally, it is noted that at high laser irradiance (of order 10^{15} W/cm^2), nonlinear inverse-Compton scattering has been observed wherein several photons interacted with one electron, producing a single-frequency-upshifted output photon (Bula et al., 1996).

2.6. High-harmonic-generation sources (strong-field science)

With the advent of sub-picosecond lasers and their high peak powers, a new regime of highly nonlinear laser–matter interactions could be explored. One of the surprising phenomena discovered was a new way of making soft X-rays with lasers via high-order harmonic generation (HHG). HHG is commonly produced when lasers with intensities \( \geq 10^{14} \text{ W/cm}^2 \) are focused onto gas jet or solid targets. Target atoms are field ionized and during a part of the cycle of the driving field, accelerated electrons emit coherent radiation of integer harmonics of the fundamental frequency with an angular divergence approaching the diffraction limit. The ponderomotive forcing of the electrons is proportional to laser intensity. As a result of improvements in the phase characteristics of the optical pulses, as well as their short durations, highly intense laser fields have created harmonic orders beyond 300, pushing this technique’s reach into the soft X-ray regime up to about a kilovolt (Brabec and Krausz, 2000). Since HHG occurs during only part of an optical cycle (one optical cycle is about 2.5 fs), high-harmonic X-ray pulses have been produced with durations of only a few hundred attoseconds (Paul et al., 2001). Further, the use of adaptive optics has been used to enhance and select individual harmonics. Finally, while the harmonic spectrum is quantized in multiples of the fundamental laser frequency, by varying gas-target pressure or the position of the focus, it is possible to tune the overall spectrum by more than several fundamentals (Umstadter, 2003). This selectability and tunability then gives the investigator access to table-top, tunable, coherent, short-pulse, soft X-rays.

One obvious application is the study of ultrafast phenomena, most commonly imaging or dynamics using pump–probe techniques. Time-domain studies have been used to observe atomic motion and femtochemistry with femtosecond laser pulses. With HHG and X-ray pulses in the attosecond regime (the natural time scale of an atomic electron is 150 as, the period of the Bohr orbit in hydrogen), one can now perform time-domain studies of electron dynamics in atomic and molecular systems that involve both valence and core transitions. One such elegant experiment (Drescher et al., 2002) used harmonic radiation at 97 eV to produce M-shell vacancies in krypton. Since HHG pulses are well synchronized with the driving field, it can be used as the pump of the Kr system, which is then probed with variable delay by the high-field fundamental. Using a ponderomotive streaking technique, the sub-femtosecond time evolution of Auger electron emission was tracked and the 3d core hole was found to have an 8 fs lifetime and delayed from the exciting X-ray pulse by a few fs.

A very special application area is now addressed with the ability to produce X-rays with the HHG characteristics outlined above that are energetically within the “water window,” that is, between the K-edge absorption energies of carbon and oxygen. In this region, X-rays are weakly absorbed in water compared to other biomaterials, permitting in vivo imaging and holography without the need for contrast agents or dehydration and attendant collapse of the biostructure to be studied (Wachsmann-Gogui et al., 1994).

3. Diagnostics and metrology

The elucidation of new scientific phenomena requires the concurrent development of a supporting infrastructure of measurement science, standard reference data, and absolutely calibrated diagnostics traceable to international units of measure. This not only helps to define the fundamental mechanisms that emerge in new interaction regimes of light and matter, but also gauges system performance as the physics is harnessed into specific applications.

Certainly a wide variety of diagnostic techniques are required to help enable the development and understanding of the exotic sources of laser-produced X-rays surveyed in this paper. We focus here on spectroscopic diagnostics. Listed below are examples of the types of information derivable from laser-produced X-ray spectra:

- identification of X-ray sources and ionization state(s) from bound–bound line transitions,
- measurement of spectroscopic line and satellite ratios (plasma temperature and opacity),
- measurement of bound–free continuum emission (presence of hyperthermal electrons),
- measurement of line widths and shapes (plasma density and source size),
- spectra as a function of time or space, and
- measurement of absolute intensities (conversion efficiency of laser light to X-rays).

So a spectroscopic X-ray diagnostic wish list would then include:

- broad spectral coverage,
- high resolving power,
- large dynamic range of detection,
- high sensitivity,
- data acquisition within very short or very long integration times,
- spatial and/or time resolution of spectra,
- high signal-to-noise ratios in hostile HED and/or strong-field environments,
- designs that accommodate point-like and/or extended sources, and
- absolute energy and sensitivity calibrations.

Most of our efforts in this field have been the design, production, calibration, and fielding of wavelength-dispersive X-ray spectrometers as a diagnostic of exotic X-ray sources. These have been fielded to help characterize the performance and X-ray spectra from advanced medical X-ray sources, laser-produced plasmas, terawatt pulsed accelerators, intense ultrafast laser sources, and inverse-Compton backscatter sources. Most recently
this has been motivated primarily by the need to better understand ns laser-matter interactions in ICF (Hudson et al., 2002, 2006; Seely et al., 2003, 2006, 2007; Section 2.1) and ps laser–matter interactions for the fast ignition and point-projection radiography applications (Tommasini et al., 2007, 2008; Park et al., 2008, Section 2.2). Ongoing investigations requiring hard X-ray diagnostics include the conversion efficiency of laser light into X-rays, the hot-electron energy distributions, and the circulation of hot electrons in laser–solid target irradiation.

One implementation of a bent crystal spectrograph (Hudson et al., 1996; Chantler et al., 1996) has been shown to be particularly well matched to the wish list outlined above; it has been shown to perform well in the noisy environments of strong-field and HED experiments (Hudson et al., 2006) while registering spectra from 12 to 160 keV. This type of diagnostic employs the Cauchois geometry, wherein the diffracting planes of a cylindrically bent crystal are normal to the crystal surface that faces the X-ray source. As shown in Fig. 1, this implementation has the advantage that it admits no line of sight from the source to detector when the source is not so extended in the plane of dispersion. In this case, only dispersed light illuminates the detector, giving a high signal-to-noise result from targets that are also emitting debris and other radiation and particles. The symmetric illumination of the crystal produces two mirror-symmetric spectra on the detector, and since the plate function is known (Hudson et al., 1996) this enables the assurance of in situ energy calibration. This geometry naturally accommodates the small diffraction angles of high-energy X-rays as well as time-integrating or -resolving detection schemes. This type of diagnostic is also well matched for characterizing much lower-power, table-top X-ray sources, such as those being developed for narrow-band medical-imaging applications, where the spectrum can be easily integrated over many pulses of the laser.

Recently a formalism was developed for the Cauchois X-ray spectrometer that quantifies the various contributions to the observed line width (Seely et al., 2008), including source size, detector spatial resolution, crystal thickness, crystal rocking curve, spectrometer aberrations, and natural line width. The two primary contributors to the registered line width are typically the detector resolution and the size of angle subtended by the source of X-rays in the plane of dispersion. To first order, extended X-ray sources are focused onto a so-called Rowland Circle (RC; shown in top of Fig. 1) with diameter equal to the radius of the crystal curvature. Hence the registered line width is usually dominated by detector resolution when the spectrum is acquired on the RC. If the source size is relatively small, it will be seen to defocus only slowly as a detector is positioned further off the RC (Hudson et al., 2007). In such cases, the source size is the dominant contributor to the measured line width; in these cases very-high instrumental resolving powers are achievable, normally challenging in hard X-ray wavelength-dispersive spectroscopy. This dependence of resolving power on detector resolution and position is shown in the two spectra of Fig. 1. These were acquired simultaneously off and on the RC from a single picosecond shot of

![Fig. 1. Demonstration of on- and off-Rowland Circle spectroscopy. The two spectra were acquired simultaneously using the geometry implemented as shown from a single picosecond shot of the LULI Laser (Laboratoire pour l’Utilisation des Lasers Intenses) focused onto a CsI target. The K lines from Cs and I are observed in both cases but with different instrumental resolving powers. The (10-11) diffraction planes were used, perpendicular to the crystal entrance surface of a quartz plate that was elastically bent to a 254 mm radius of curvature.](image)
the LULI Laser (Laboratoire pour l’Utilisation des Lasers Intenses, 100 J, 10 μm focal spot, 10 TW/cm²) using a CsI target. This is shown here for illustration; detailed analysis of these experiments is being published elsewhere (Seely et al., 2009).

4. Summary

This paper surveys the recent advances in laser science that can be applied to produce novel sources of X-rays and enhanced X-ray applications. The ability to now sculpt laser pulses in time and space and deliver large photon densities to gaseous and solid targets can lead to a wide variety of absorption mechanisms, some highly nonlinear and relativistic. These techniques create extreme states of light and matter, which are better understood and controlled with feedback from advanced X-ray diagnostics. These enabling technologies will facilitate this emerging marriage between the laser and X-ray sciences and will give birth to a wide array of applications, many of which are not yet dreamt of.

References


