Advancements and Applications of Laser-Produced Plasmas

Daniel Ross Witteman

ABSTRACT
The concept of a laser blaster turning an enemy into a cloud of vapor may seem purely fictional, but the science that inspires this idea has real-world applications. Strong, focused lasers can indeed melt, vaporize, and ionize nearly all forms of matter. The highly energetic and dynamic characteristics of ionized gases, or plasmas, have been exploited to address many technical challenges that humanity faces today.

Presented here is a brief review of the physics behind lasers and the plasmas they can generate, followed by several applications of laser-produced plasmas that are critical for the future of our environment, technology, and energy. Using lasers to ionize materials has proven useful in measuring and remediating contamination, manufacturing advanced computer chips, and performing groundbreaking research in fusion energy.

Keywords: Laser-produced plasmas, Environmental remediation, Extreme ultraviolet lithography, Inertial confinement fusion, Laser-induced breakdown spectroscopy

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Introduction

The invention of the laser has opened a broad avenue of scientific research and industrial applications ranging from fundamental physics, material processing,[1] medicine,[2] dentistry,[3] and communication systems.[4] Lasers possess a refined ability to deliver enormous amounts of photon energy to miniscule targets in a short period. One significant area where these properties are crucial is in the generation of plasmas, a highly energetic state of matter. Plasmas contain the charged particles of ions and unbound electrons which exhibit diverse properties. This review article will cover the fundamental concepts of laser-produced plasmas (LPP) and the forefronts of their applications such as environmental monitoring[5] and remediation,[6] the commercial production of today's most advanced semiconductors,[7] and the effort to understand and harness controlled nuclear fusion.[8]

In addition to being relevant in the critical areas of environment, economy, and energy, the applications selected for this review display distinct regimes of plasma dynamics. Each application is presented here in order of increasing plasma temperature. The relative complexity of each application also increases with the engineering of a higher-temperature plasma, providing a view of the technological range of the field of LPP. While this is by no means a comprehensive set of the applications of LPP, the review intends to highlight some of the most advanced and currently relevant use cases. Pollution and contamination of air, water, and soil are issues faced by nearly all industrialized countries. The rise of artificial intelligence has placed increased demand on highly capable semiconductor chips. The ever-growing energy needs of modern societies necessitate a breakthrough in producing clean, carbon-free energy. The plasmas produced by intense laser radiation play a role in addressing all of these challenges.

Fundamental Physics of Lasers and Plasmas

From the common red laser pointer a child might play with to the powerful invisible beams capable of downing a drone, lasers are an inherently quantum phenomenon. The theoretical framework that underpins the operation of lasers was first predicted by Albert Einstein's quantum theory of radiation.[9] Light is described as massless, wavelike particles called photons carrying energy $E = hf$, where h is the universal Planck's constant and f is the frequency of light such that $\lambda = c/f$ is the wavelength of the photon where c is the speed of light in vacuum. A photon of a given wavelength thus has the energy $E_{\text{photon}} = hc/\lambda$. The discrete energy of a photon can be absorbed and emitted by electrons transitioning between different energetic states and also by molecules transitioning between vibrational states. These energetic transitions are quantized to specific values due to the atomic or molecular parameters.

Lasers rely on stimulated emission, where a photon incident on an electron in an already excited energy state E_{Hi} has a probability of causing the electron to descend into a lower energy state E_{Low}, releasing the energy as an additional photon $E_{\text{photon}} = E_{Hi} - E_{Low}$ with an identical wavelength and direction as the incident photon.[10] If these two photons continue to propagate through a medium with the same excited particles, a so-called gain medium, they will harvest more photons and form a laser beam as shown in Figure 1A. The distinct properties of lasers are outlined in Table 1. The population of excited states depletes as the beam propagates through it, so the gain medium must be pumped with energy using high voltages or flash lamps depending on the type of medium and the targeted lasing transition. The first laser was built in 1960 from a ruby rod pumped with flash lamps to emit a visible red beam.[11] Now many laser infrastructures exist at a variety of wavelengths, from the microwave and infrared to visible and ultraviolet. Extremely high power levels are reached by triggering a short pulse through a gain medium that is highly saturated with excited states.

Generating a plasma from neutral matter involves rapid and intense heating of the target. When any light encounters matter, some percentage of the photons are absorbed, exciting the electrons into higher energetic states, while the remaining photons are either transmitted or reflected. The relative fraction for each of these processes depends on the material's properties and the light's wavelength and intensity. When the energy absorbed by an atom exceeds the binding energy

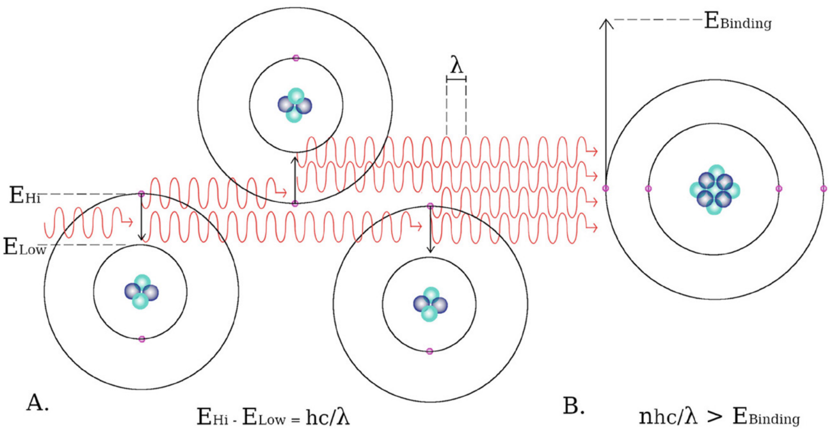

Fig 1 | Fundamental physics of (A) laser propagation and (B) ionization by multiple photon absorption

Table 1 | Properties of laser light

Monochromaticity	The light emitted by a laser is tightly concentrated around a single wavelength corresponding to the energy transition of the gain medium
Collumnation	Photons travel in the same direction, so the beam can travel long distances with minimal spreading
Coherence	The wavefronts of light are in phase in both space and time such that their amplitudes add together for a more intense beam
Tunability	The beam shape can be manipulated spatially and temporally to create short or long pulses with wide or narrow focus

Table 2 | Processes in plasmas

Process	Symbolic	Reverse Process
Photoexcitation An atom (*M*) absorbs one or (*n*) several photons (*γ*) of wavelength *λ* raising an electron within the atom to an excited state (M^*)	$n\gamma + M \rightleftarrows M^*$ $n\,hc/\lambda < E_{Binding}$	**Spontaneous emission** Excited electron decays to a lower energy state with the emission of a photon
Photoionization One or several photons of wavelength *λ* have sufficient energy to create an ion (M^+) and a free electron (e^-)	$n\gamma + M \rightleftarrows e^- + M^+$ $n\,hc/\lambda \geq E_{Binding}$	**Radiative recombination** Electron and ion recombine with the emission of a photon
Collisional excitation Free electron collides with an atom transferring kinetic energy to the atomic system	$e^- + M \rightleftarrows e^- + M^*$	**Collisional de-excitation** Electron inelastically collides with an excited atom, gaining kinetic energy
Collisional ionization Free electron collides with atom transferring enough energy to release an additional electron	$e^- + M \rightleftarrows 2e^- + M^+$	**3 body recombination** Two electrons collide with an ion and one is bound
Bremsstrahlung Free electron is slowed and its kinetic energy is released as a photon in a continuum of energies	$e^- + M^+ \rightleftarrows e^- + M^+ + \gamma$	**Inverse bremsstrahlung** The absorption of a photon by a free electron increases its kinetic energy

$E_{Binding}$, an ion and an electron are produced. Electron binding energies are typically a few to tens of electron volts (eV) for outer shell electrons and up to thousands of eV for inner shell electrons. The photon energies of many commercially available lasers (i.e., ~1.17 eV for Nd:YAG lasers at 1064 nm) are typically below the threshold of ionization of many atoms; however, laser pulses with intensities higher than 10^8 W/cm^2 are generally able to ionize all states of matter via several mechanisms described in Table 2. The absorption of a photon or simultaneous absorption of *n* number of photons can lead to ionization if $n\frac{hc}{\lambda} \geq E_{Binding}$.[12] The process of multiple photon ionization is depicted in Figure 1B, where an atom absorbs four photons to liberate an electron.

LPP are highly dynamic transient systems. The rapid absorption of laser energy by a target leads to vaporization of solids and liquids and a shockwave of ions expanding out from the focal spot of the laser. Laser energy is directly absorbed through inverse bremsstrahlung, accelerating electrons to high energies.[13] Fast free electrons create a cascade of ionization in collisions with neutral or excited species.[12] Excitations below the ionization threshold still contribute to plasma growth by raising electrons into higher energy levels that have lower binding energies.[14] The average kinetic energy of the electrons, also known as the electron temperature, determines many properties of the plasma. Ions typically have a lower kinetic energy due to their higher mass, but they still contribute to the overall plasma temperature. After the laser pulse terminates, the expanding plasma cools through collisions, radiative recombination, and bremsstrahlung emission. The cooling plasma emits both light at distinct wavelengths and in a continuum due to these processes.[13] The radiative properties of LPP have important implications in the applications discussed below.

LPPs in Environmental Remediation

A high cost of industrialization is that a litany of pollutants ranging from heavy elements[15] to toxic chemicals with long lifetimes[16,17] are constantly entering our air, water, and soil. There are concerted efforts by governments and nongovernmental organizations around the world to monitor levels of pollutants and remediate them to prevent damage to humans and other organisms.[18] One major growing concern is the widespread presence of "forever chemicals" like per- and polyfluoroalkyl substances,[17] which have rapidly spread in biological systems and are likely carcinogenic.[19] LPPs are increasingly being used as a monitoring tool to detect both the level of contamination of certain contaminants[20] and also an indirect means of remediation.[21]

Contaminant Monitoring

The method of laser-induced breakdown spectroscopy (LIBS) utilizes short, intense laser pulses to heat a small portion of the sample to tens of thousands degrees K, which is around 1 to several eV.[22] Each element within the sample emits light at distinct wavelengths or "spectral lines," such that analyzing the light from the plasma allows for the detection of the atomic species within (Figure 2). This monitoring method often requires no sample preparation and is fast, and relatively economical.[23,24] When preparation is required, it can be as simple as grinding a heterogeneous solid and then pressing it into a pellet or freezing a liquid sample for an enhanced emission spectrum.[25] Firing a second laser pulse into the plasmas also strongly enhances the emission spectrum, especially for samples that are immersed underwater.[26–28]

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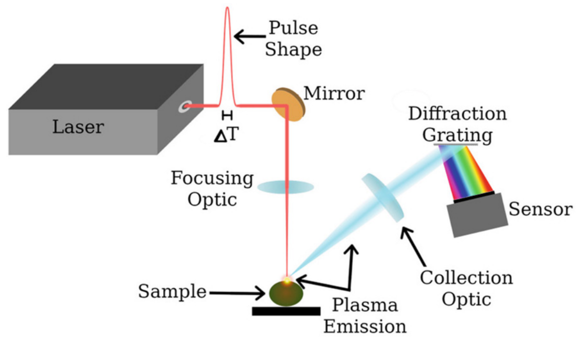

Fig 2 | Laser-induced breakdown spectroscopy

The ease or lack of preparation in LIBS allows for faster monitoring of contamination and can enable a more thorough analysis of the migration of contaminants than would be economically feasible with other methods. Senesi et al. demonstrated that LIBS can be used to track heavy metal contamination from the soil into the roots and shoots of a plant used for bioremediation.[24] They finely ground the soil and plant samples and pressed them into pellets. The pellets were placed in a rotating target holder to take multiple measurements of one sample, each on a newly ablated surface to assist reproducibility. When compared to the conventional method of measuring the spectrum of an inductively coupled plasma, LIBS was found in good agreement, although the uncertainty was higher due to the smaller sampling volume.[24]

There are some notable limitations of the LIBS sampling method, some of which can be mitigated with adequate sample preparation and experimental setup. The laser's tight focus has the benefit of being minimally invasive, but this can also lead to sampling error in bulk and heterogeneous materials because only a small amount is sampled. The formation of deep craters due to continuous ablation can lead to the enrichment of certain elements over others within the sample.[5] When possible, rotating the material while sampling can mitigate these errors. The spectrum obtained from a LIBS is highly dependent on the laser parameters (wavelength, energy, and pulse profile), the sample grain size and stoichiometry, and the background gas composition and pressure.[29] Understanding the plasma dynamics, such as the relative abundance of excited species and their ionization degrees, gives insight into selecting the most optimal spectral lines for LIBS analysis. Machine learning-based approaches could be applied here to provide real-time feedback on the plasma parameter for improved measurement accuracy and consistency.[30]

Remediation

In addition to helping us understand the specific pollutants contaminating an environment, LPP can also play a significant role in breaking down and removing certain toxic and dangerous substances. Laser breakdown generates reactive oxides and nitrides in air as well as hydroxyl radicals in water. These reactive species are effectively able to break down dyes, microbial contaminants and potentially PFAS.[31–33] Lower-temperature plasma sources produced from high-voltage discharge are currently favored for plasma remediation applications as they are more energy efficient in generating reactive species.

LPP can still play a role in remediation with the generation of nanoparticles and nanocomposites in their ablated plasma plume.[21] These particles with sizes of less than 100 nm have distinct properties from bulk material and effectively remediate various pollutants due to their high reactivity and absorption capabilities.[34,35] Varying the laser parameters such as the pulse length and wavelength can even enable control over the size distribution of the generated nanoparticles. Scaling the nanoparticle size changes their effective surface area, an important factor that affects their ability to remediate.[36] Chakravarty et al. demonstrated that lengthening the pulse duration of a 30 mJ ultrashort pulse laser shifted the size distributions larger for both silver and copper nanoparticle.[37] Metal nanoparticles and their composites can play an important role in the degradation of hazardous substances through catalytic reactions.[6] High capital and energy costs from purchasing and operating laser equipment still hinder the adoption of LPP for remediation.

Extreme Ultraviolet Semiconductor Lithography

Since the early computers of the 1960s, the number of transistors on integrated circuits has increased exponentially, doubling approximately every 2 years. This rate of progress is known as Moore's law. Modern chips are manufactured through photolithography, a process where a silicon wafer is imprinted with intricate patterns through the use of light and photosensitive chemicals to create a network of transistors. To increase computing power, it is essential to enhance transistor density. However, the wavelength of the light used to expose the wafer acts as a limiting factor, which can be described by the formula: Resolution = $k\lambda/NA$, where k is an empirical parameter, λ is wavelength, and NA is the numerical aperture.[38] To resolve the increasingly smaller features on advanced chips, there is a demand for short-wavelength light and, consequently, high-energy photons.

Research into the generation of soft x-rays or extreme ultraviolet (EUV) light, which falls within the range of 10–30 nm, has been under development since the 1980s.[38–40] These early works used synchrotron radiation as a soft x-ray source, as well as LPP from gold targets. Other sources of EUV include the sun, free electron lasers, and ultrashort pulsed laser plasmas. Highly ionized heavy metals have shown capabilities for emitting EUV, with tin ultimately chosen as the fuel source for future lithography due to its emission peak at 13.5 nm (~92 eV) under intense laser irradiation.[41] The short wavelength of EUV is absorbed by most materials, including air and conventional optics. However, multilayer mirrors of Si and Mo are capable of reflecting EUV with up to 70% efficiency. Since the imaging system of a lithography scanner implements several mirrors, achieving a high source power of

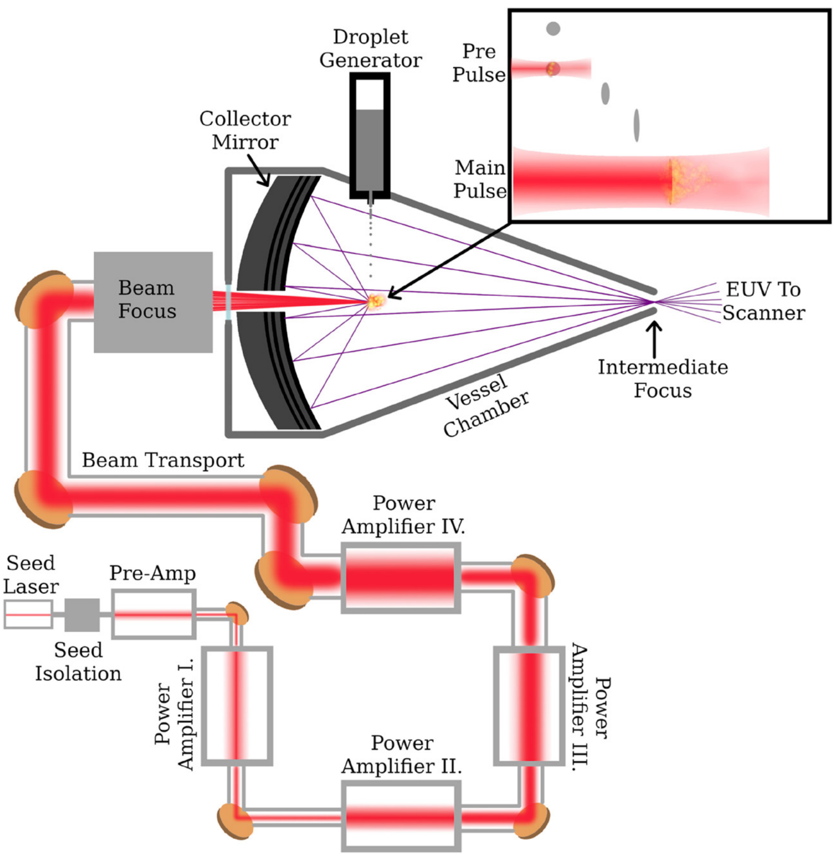

Fig 3 | Laser-produced plasma EUV light source, with an inset showing the laser droplet interaction at the primary focus of the collector mirror

>250 W is required to deliver sufficient EUV to the wafer. Accomplishing this has been a monumental feat of engineering.

The commercial-scale EUV source was developed by CYMER,[42] a company later purchased by ASML.[43] The source is a laser produced tin plasma that is pulsed at a high frequency with the EUV reflected by a large ellipsoid multilayer collector mirror as depicted in Figure 3. Tin is injected by a droplet generator using high-pressure inert gas applied to a melted tin reservoir, forcing a stream of microdroplets out of a small orifice. The spherical droplets fly through the vessel, passing through metrology lasers and cameras to precisely position the droplets and fire the drive laser at the primary focus of the collector mirror. The drive laser is a CO_2 gas laser operating at 10.6 µm that passes through a preamplifier and then four high-powered amplifiers to achieve an average output power of 30 kW. A "prepulse" first flattens the droplet into a thin disk to improve energy coupling for the main pulse, which heats the tin to plasma temperatures sufficient for EUV generation.[44] The prepulse also prevents excessive generation of macroparticle debris that can implant on the collector mirror. H_2 gas is also flown through the vessel to interact with tin ions and carry them out of the vessel to preserve collector reflectivity.[45]

When a single droplet target is heated by ~600 mJ of laser energy, the plasma reaches electron temperatures of 30–40 eV[46] with tin ions reaching ionization states of Sn^{10+} – Sn^{15+}. The plasma emits light in a broad spectrum in all directions, but the commercial process is only concerned with the emission of in-band 13.5 nm EUV directed back at the collector mirror, which amounts to ~5 mJ per pulse. To achieve the high EUV power requirements of ~250 W needed for semiconductor production, this process is scaled to a high repetition rate of 50 kHz. Stability must be maintained in droplet size, spacing, and positioning, as well as in laser power, pointing, and timing, in order to deliver EUV within the dose requirements for semiconductor production.[45,47]

Research is underway to incrementally improve EUV output and conversion efficiency. This is accomplished with a separate 1 µm laser as the prepulse to form the target.[48,49] Removal of the CO_2 prepulse prevents a back-reflection from the droplet from entering the power amplifiers, negating a parasitic gain–loss mechanism.[50] Laser gain is then reserved for use by the main pulse to heat the plasma.[48] Conversion efficiency is improved in the way the short pulse and shorter wavelength of the 1 µm laser interact with the liquid droplet and more effectively form a wide target to be heated by the main pulse.[51,52] Scaling the repetition beyond 50 kHz further multiplies the EUV power increases. With these methods and additional laser amplification, research sources have achieved stable EUV output at 600 W.[48]

While the CO_2 gas laser was chosen for its industrial robustness at high repetition rates, these systems are notoriously inefficient. The development of efficient high-power diode lasers capable of firing at high repetition rates could drastically reduce the energy consumption of EUV systems. Incorporating magnetic confinement of the plasma would further improve the plasma heating and EUV conversion efficiency.[53] Magnetic fields can also aid in debris mitigation by trapping low-energy ions.[54,55] The development of lasers that generate coherent, directional EUV could offer even more dramatic improvements by avoiding the inherent inefficiencies of mirror losses from the collector and through the scanner. Such EUV lasers are possible through high harmonic generation[56,57] and free electron lasers.[58]

Laser-Driven Inertial Confinement Fusion

The quest for controlled nuclear fusion has the potential to provide a nearly limitless source of clean and renewable energy. The fuel for the reaction is the heavy isotopes of hydrogen: deuterium (D), containing one proton and one neutron, and tritium (T), containing two neutrons. D_2 is a naturally occurring stable isotope with an abundance of 1 D for every 6700 H in natural water[59] and is easily extracted.[60] Tritium is unstable and must be generated from neutron capture: $D + n \longrightarrow T$.[61,62] The conditions for fusion require that the fuel has a combination of high temperature and density to overcome the mutual repulsion of positive nuclei, binding them into helium and a fast neutron: $D + T \longrightarrow \text{He}\,[3.5\text{ MeV}] + n[14.1\text{ MeV}]$. The high-energy neutrons escape the plasma and carry their energy off to the chamber walls, which can serve as a tritium breeding mechanism for future reactors. For the reaction to produce net energy, the released alpha particles

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(He) must make sufficient collisions within the plasma such that they add energy to the fuel and drive further fusion events.

The Lawson criteria for ignition describes when the self-heating from alpha particles exceeds the energy input and cooling mechanisms.[63] The laser's ability to generate hot, high-density plasmas has made them a prime candidate for heating and compressing fusion fuels to high densities needed to stop alpha particles within the plasma. The concept has been in development since the 1970s[64] with recent breakthroughs in generating self-heating fusion output.[65]

The infrastructure to generate inertial confinement fusion can be as simple as two opposing beams on a DT target fired simultaneously to generate an implosion of the fuel;[64] however, modern designs developed by the National Ignition Facility (NIF) have become increasingly more intricate by using 192 lasers to illuminate the target with a higher degree of symmetry.[66] The approach that has finally achieved Lawson's criteria for ignition uses an indirect drive pictured in Figure 4, where the lasers couple energy to an outer shell called a hohlraum—a high atomic number material that emits a bath of x-rays. An inner capsule within the hohlraum houses the frozen and gaseous DT fuel within a thin lining material. The x-rays ablate the capsule lining inward and implode the fuel at a high velocity to generate a central hot spot where fusion begins and continues heating the surrounding fuel. Key developments in achieving this feat include achieving an extremely high-energy laser pulse with a tailored temporal profile to drive compression and engineering the hohlraum and capsule materials and geometries to efficiently absorb laser energy and mitigate instabilities that lead to loss of compression and cooling of the plasma.[67]

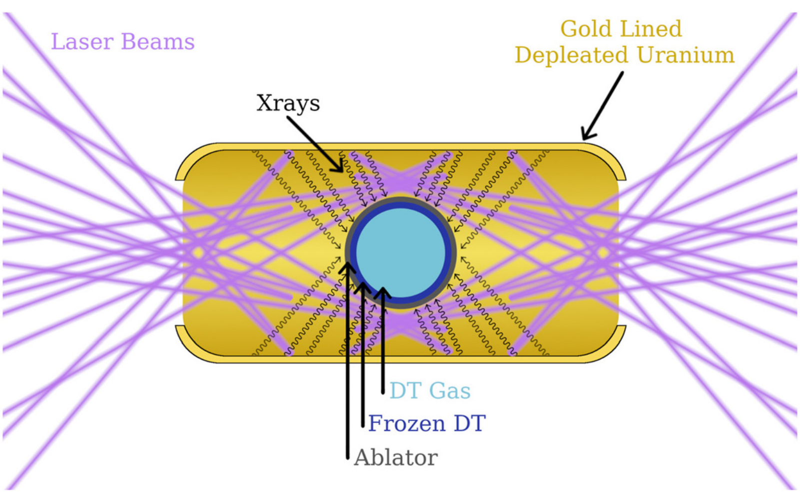

Fig 4 | Indirect drive laser inertial confinement fusion

The NIF laser system and hohlraum upconvert the frequency of the photons several times before they are finally delivered to the DT target, as depicted in Figure 5. Beginning with a single nanojoule-level infrared pulse at 1053 nm, the beam is split several times and sent through a series of glass amplifiers to reach a pulse energy of 4 MJ. Near the target chamber, the infrared beams are frequency tripled by passing through crystals of potassium dideuterium phosphate to reach an ultraviolet wavelength of 351 nm. The frequency conversion incurs a ~50% loss of the beam's power, but the higher photon energies of the UV beams are more effective at coupling their energy to the hohlraum.[68,69] In recent experiments, 2.05 MJ of UV laser energy has heated the hohlraum to a peak radiation temperature of 313 eV, which emits soft x-rays in the EUV range. The fusion output has been measured as high as 3.88 MJ in a shot cataloged as N230729[70] for a gain of energy approaching 2. The fusion reaction reheated the hohlraum far past the initial peak to nearly 350 eV, which is over 4 million celsius. Although this is a significant feat in the long history of fusion research, further developments are required to enable fusion as a viable energy source. The energy required to actually generate and amplify the laser pulse still eclipses the fusion output at over 300–400 MJ.[71,72] The fusion output will need to increase by a factor of over 100 to achieve true energy breakeven. Imposing a magnetic field along the hohlraum has been shown to increase hotspot temperatures, although these experiments must be conducted with constraints of a lower laser energy of 1 MJ and room temperature fuel that limits total fusion yield.[73] Further exploration of laser-driven fusion within magnetic fields could lead

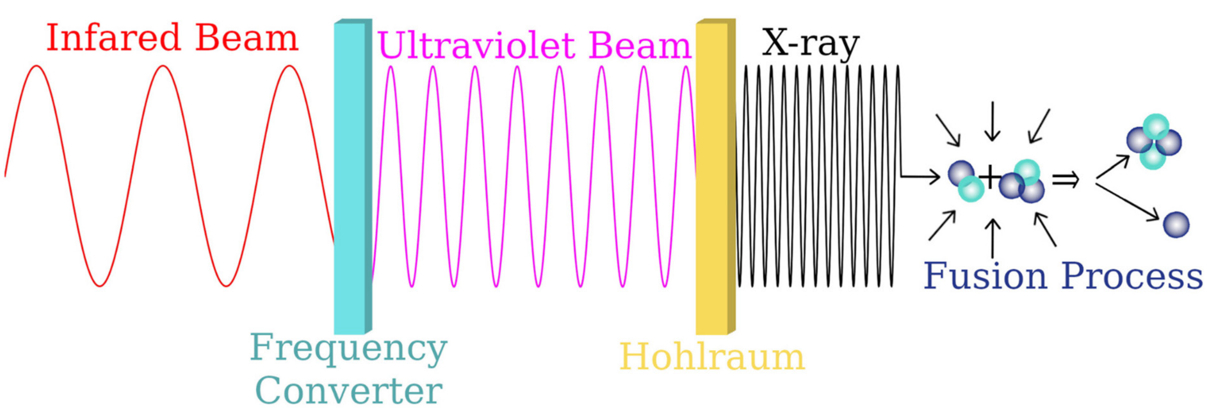

Fig 5 | Photon frequency conversion from infrared to ultraviolet to x-ray driving fusion to more breakthroughs in achieving the high fusion gain needed for future reactors.

Conclusion

The utility of the laser and the plasmas it creates has far-reaching applications across modern society. There are still many other important applications of LPP that have not been covered here: pulsed laser deposition for thin film processing has implications for efficient energy materials, x-ray generation for medical imaging, and laser pair production for antimatter research, to name a few. Converting the energy of an excited gain medium into a concentrated light beam underpins the laser's operations. In applications, laser photons are converted into a different form of desired energy or emission within the plasma. Research in different gain mediums and laser infrastructures has led to the high-energy and short pulses that drive many LPP applications. Expanding the available wavelength selection of lasers could also lead to more novel applications. For environmental applications, LPP serves as an effective tool with modest infrastructure requirements for both monitoring levels of contamination and remediating them. Combining laser-assisted remediation methods with real-time monitoring of contamination levels through LIBS could be an effective future avenue for cleaning up highly contaminated waste streams.

Applications targeting higher-temperature plasmas require higher laser energy input. In lithography, the EUV emitting plasma from firing multiple lasers on tin droplets is an operational tool for printing extremely small features on advanced chips. In fusion research, symmetric laser illumination heats a small target to unleash an x-ray bath that rockets a fuel capsule into itself in an effort to release even more energy. The addition of a magnetic field in both of these applications offers a means of increasing the plasma heating efficiency. From the seed laser pulse, through power amplification, and onto target heating there are loss mechanisms in each energy conversion step that should be understood and minimized to improve efficiency. Powering laser infrastructure with renewable energy generation would offset their high energy demand, and there are even prospects of using focused sunlight as a laser pump.[74] Further improvement in laser efficiency and plasma production would lead to a smaller environmental footprint of LPP technologies and expand the feasibility of laser-based solutions.

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