Plasmas Produce Very Intense X-ray Pulses

With such bright sources, researchers can record a diffraction pattern or absorption spectrum with just one 10^{-9} second shot

If scientists using x-rays have a dream, it must be of an x-ray laser. Although immediate prospects for such a laser are slight, an offshoot of laser fusion research may provide the next best thing. A very intense pulse of laser light striking a target produces a hot plasma near the vaporized surface. The plasma, in turn, emits a burst of x-rays powerful enough to allow diffraction patterns or absorption spectra to be obtained from just a single shot lasting about 10^{-9} second. This capability opens the way to the study of very short-lived structures, such as intermediate species in chemical reactions. Another use is in x-ray lithography, an experimental technique for the replication of finer patterns in microelectronic circuits than now common, and in the related field of x-ray microscopy.

The size and cost of lasers of the type needed to make plasmas are much less than those of the electron storage rings used to produce synchrotron radiation. It is possible therefore that plasma x-ray sources will permit researchers to do, in their own or in nearby laboratories, the kinds of experiments which now require them to travel to distant synchrotron radiation centers. At least one group has constructed a source in which an electron beam, rather than a laser, is used to create the plasma; this source is much less expensive to build than even the laser-induced plasma sources.

Anybody who was a younger brother or sister probably knows what it is like to wear hand-me-down clothes. Somewhat the same sort of situation applies to laser-induced plasmas and laser fusion. The largest laser fusion program is the one at the Lawrence Livermore Laboratory, which is about an hour's drive east of San Francisco. In the space of 5 years, researchers at Livermore have progressed from a large solid state (glass) laser that could produce pulses with a peak power of about 200 billion watts to a monstrous system comprising 20 separate beams that puts out a hundred times more power. Already under construction is an even larger giant that will be ten times more powerful—200 trillion watts. The question is what to do with the smaller lasers left over. Much fundamental physics related to laser fusion can still be done with these "hand-me-downs," according to Moshe Lubin, director of the University of Rochester's Laboratory for Laser Energetics, where a 24-beam glass laser comparable to Livermore's present one is nearing completion. But the smaller lasers have also become available for other types of experiments.

A second tie-in to laser fusion comes from the use of x-rays as a diagnostic tool to analyze the degree to which a pulse of laser light can compress and heat the tiny glass Microballoons filled with deuterium and tritium that will serve as the fuel in future laser fusion reactors. As a result, says Nat Ceglio of Livermore, researchers have spent much time studying the properties of the x-rays produced during laser irradiation of the Microballoons and the relation between these properties and the characteristics of the laser pulse (energy, duration, wavelength, and so on) and the nature of the target it strikes. At the same time, researchers also were looking for targets other than Microballoons that might maximize the production of x-rays, which could then be used for other purposes, such as spectroscopy or diffraction experiments. Much of the work on nonfusion applications of plasma x-ray sources has been by groups at the Naval Research Laboratory (NRL) and the Battelle Columbus Laboratories, where funding to build the largest lasers was unavailable.

But it is only within the last year that applications of hot plasmas as intense x-ray sources have been demonstrated. The most recent is the use of a laser-induced plasma to generate extended x-ray absorption fine structure (EXAFS) spectra by Philip Mallozzi and his coworkers at Battelle (*Science*, in press). EXAFS refers to the oscillations or wiggles in the absorption spectra at energies up to several hundred electron volts above an x-ray absorption edge. Analysis of the wiggles yields information about the radial distance between the absorbing atom and its nearby neighbors and the number of neighbors in solids, liquids, and gases. Because the fine structure is so small, however, it can take spectroscopists days or even weeks to get statistically significant data with conventional x-ray sources. Thus the EXAFS technique really became popular only when synchrotron radiation facilities that could produce high fluxes of x-rays became available a few years back. With synchrotron radiation, spectra of high quality could be taken in minutes on ordinary (dense) materials, and recording spectra on samples once considered too dilute to study became a common occurrence. But the Battelle group took an EXAFS spectrum of aluminum with just one pulse lasting 3×10^{-9} second.

To accomplish this feat, according to Mallozzi, the investigators used a glass laser capable of emitting infrared pulses (1.06 micrometers) containing about 100 joules of energy, which, with the indicated pulse length, corresponds to a peak power of 33 billion watts. The laser light was focused into a spot, 100 to 200 micrometers in diameter, on the surface of an iron slab, the point from which the resulting x-ray pulses emanated. The x-rays then passed through an aluminum foil sample. The transmitted x-rays were dispersed (separated according to their wavelength) by a flat crystal monochromator onto a photographic film, which recorded the intensity of each transmitted wavelength.

Earlier this year, an altogether different use of a laser-induced plasma source was reported by Robert Frankel and James Forsyth, of Rochester's laser laboratory, who have done low-angle x-ray diffraction studies of several biological macromolecules (*Science*, 11 May 1979, p. 622). Low-angle diffraction, which does not require crystalline specimens, is used to study ordered structures whose characteristic periodicity may be from tens to hundreds of angstroms as compared to the repeat spacings of a few angstroms in the crystalline structures probed by conventional x-ray diffraction.

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Frankel and Forsyth used the so-called glass development laser at Rochester; this is the laser that served as the prototype for the laboratory's present six-beam Zeta laser. The investigators focused a pulse containing about 90 joules of energy and lasting about 4×10^{-10} second into a 100-micrometer-diameter spot on a Saran target. The spot was immediately vaporized and, among other things, produced chlorine ions (Cl^{15+}), which emitted x-rays at an energy of 2790 electron volts (4.45 angstroms). Each pulse contains about 8×10^{13} x-ray photons, enough to allow recording of a diffraction pattern in one shot, says Forsyth, when the x-rays are focused onto the sample and when a sensitive image intensifier camera is used.

What is significant about these two results is obviously not that one can now record x-ray absorption or diffraction data in a fraction of a second—nobody would have the time to analyze all the information coming from experiments done at such a rate. The point, agree Mallozzi and Forsyth, is that it is now possible to freeze in time species that are very short-lived or have rapidly changing structures because the duration of the x-ray pulse is comparable to that of the laser pulse, namely about 10^{-9} second. In particular, it should eventually be feasible to make "movies" of dynamic effects, such as the change in the structure of a contracting muscle. For example, the muscle could be stimulated and the laser-induced plasma source would be triggered to flash at intervals of a few times 10^{-9} second or any longer spacing. Because the big glass lasers absorb so much heat as each pulse is produced, it takes up to several hours to cool enough for the next shot. Thus, the evolution with time of structures can now be followed only by waiting for the laser to cool between each frame. However, there may be ways to overcome this difficulty, such as breaking one large laser pulse into several smaller ones, according to Mallozzi.

Apart from the short length of the pulse, the feature of laser-induced plasma sources that makes such stop-action experiments possible is the intensity of the x-rays produced. Synchrotron radiation sources are the nearest competitor in terms of intensity. Synchrotron radiation also comes in pulses, which can be as short as those from plasma sources, but the number of x-ray photons in each pulse is several orders of magnitude less, at least for the present. However, points out Peter Eisenberger of Bell Laboratories, ways to enhance the output of x-rays from existing synchrotron radiation centers are on the way, as are all new facilities designed to optimize x-ray production; hence the present gap is not guaranteed to remain.

There is one way in which the two types of x-ray sources will remain irrevocably different—that is in their spectra. The synchrotron radiation spectrum is smooth and continuous with photons at every energy from the visible through the x-ray, although there is a definite maximum near the x-ray end. Plasma sources, however, bear more of a resemblance to the traditional x-ray tube spectrum which consists of a smooth continuum together with a few discrete lines of much higher intensity. Ceglio at Livermore says that the continuous portion of the plasma source spectrum and the discrete lines can be somewhat independently engineered by proper choice of the laser target and the laser pulse characteristics. In this way, as has been extensively demonstrated at Livermore in fusion-related experiments, researchers could choose to emphasize the continuum or specific lines as appropriate to the application. For EXAFS, it is the continuous portion of the plasma spectrum that is of interest in contrast to the discrete lines used for diffraction patterns obtained at a single energy.

Some observers, including Gordon Knapp of Argonne National Laboratory, are concerned that the presence of any discrete lines will interfere with the sometimes weak signals of EXAFS. Mallozzi believes, however, that by properly matching the target material to the sample, it is possible to make a source with a smooth spectrum in the energy interval desired.

One limitation of plasma sources is that the intensity of x-rays falls off drastically with increasing energy unless the temperature of the plasma is also raised when a more powerful laser is used. With 100-joule glass lasers it is not now possible to produce high fluxes of "hard" x-rays, according to David Nagel of NRL. (X-rays with an energy of greater than 10,000 electron volts are hard, according to the jargon, while those with an energy of less than 1,000 electron volts are soft. Somewhere in between is the dividing line.) Thus, without more powerful lasers or improvements in the ones used today that would increase the power delivered to the target, such as shortening the pulse length or focusing the beam to a smaller diameter, researchers cannot take EXAFS spectra of elements with atomic numbers greater than 40 nor can they do conventional x-ray diffraction with plasma sources and still retain the advantages of high-intensity and stop-action capability.

One use of plasma x-ray sources for

Glass laser at the Battelle Columbus Laboratories. The laser pulse originates in an oscillator at the rear of the room. Its energy is boosted each time the pulse passes through one of the amplifiers, which are the rectangular objects on top of the optical bench. Beneath the bench are pumps for the cooling water for the flash lamps that excite the amplifiers. In this way a pulse containing 100 joules of energy and lasting about 3×10^{-9} second can be generated. [Source: P. J. Mallozzi, Battelle Columbus Laboratories] which soft x-rays are an advantage is that of x-ray lithography, as shown by Mallozzi's group at Battelle and by Nagel and his co-workers at NRL in collaboration with Martin C. Peckarar of the Westinghouse Electric Corporation, Baltimore. At present microelectronics manufacturers shine a light, having a wavelength (4000 angstroms) on the borderline between the visible and the ultraviolet, through a metal mask to form the patterns that constitute the circuit. The use of x-rays permits patterns with features smaller than the 30,000 angstroms typical of today's microcircuits to be replicated because diffraction effects ultimately limit the size of the feature to the wavelength of the light used. In practice, other effects raise the minimum future size to a few wavelengths. One, but not the only, problem with x-ray lithography, as the process is called, is the lack of a bright enough source. A dim source means that exposure times are too long for x-ray lithography to be commercially economical.

Last year, the NRL and Westinghouse group demonstrated the ability to replicate features as fine as 7500 angstroms with x-rays from a laser-induced plasma. (This result is far short of the demonstrated ability of x-ray lithography to reproduce small features and should be construed as a proof-of-principle experiment.) Again, a glass laser was used, this time producing pulses having an energy of about 100 joules and a pulse length of about 40×10^{-9} second. The laser light was focused onto an aluminum target. X-rays from the target then passed through a filter and a mask before exposing a photosensitive polymer. However, because the system was not optimized for the lithography application (in particular, the pulse length was too long for efficient heating of the plasma) many shots were necessary. The investigators concluded that it would be possible to achieve single-shot exposures.

The reason that soft x-rays are advantageous is at least threefold. First, hard x-rays can penetrate the photosensitive polymer into the electrically active semiconductor beneath, where they can cause defects that alter the characteristics of the circuit. The fact that the x-rays are so penetrating also means that hard x-ray sources are inefficient in the sense that most of the x-rays produced are not used. Finally, those hard x-rays that are absorbed in the polymer tend to be absorbed uniformly and not according to the pattern in the mask (that is, they penetrate the mask) and thereby reduce the contrast in the pattern formed by the softer x-rays.

Conventional x-ray tubes are not only less intense than the plasma source but they also generate a higher ratio of hard to soft x-rays. Synchrotron radiation sources can be adjusted to emit as soft a spectrum as plasma sources and are at least being considered for future industrial use. However, the electron storage rings (a kind of accelerator) that produce a synchrotron radiation are expensive to build. Several million dollars has been suggested as the cost of a ring that could emit soft x-rays for lithography. Lasers that could generate hot plasmas are expected to be considerably cheaper.

Nevertheless, glass lasers capable of putting out pulses with 100 joules of energy are not inexpensive items themselves, nor are they physically small. They consist of an oscillator (laser) that starts a pulse on its way and a succession of increasingly larger amplifiers (also glass) that boost the pulse energy. The total system can fill a laboratory room and cost a few hundred thousand dollars to build. Even though such lasers are up to one hundred times less expensive than storage rings, as Edward Stern of the University of Washington points out, they are not the sort of thing every university laboratory will be able to afford.

Lasers could become cheaper, however, because the ideal laser for generating plasmas may not be the glass lasers now in use but some yet to be developed variety, says Ceglio at Livermore. Apart from the necessity of waiting for the glass laser to cool between firings, a disadvantage is that its infrared wavelength is not ideal for producing hot plasmas because not all the light is absorbed by the target at the high intensities needed to heat the plasma. Among other benefits, a better coupling between the laser light and the target would mean that a lower power laser could be used, which would be cheaper.

As one example of what might soon transpire, Ceglio suggests the possibility of an improved version of the increasingly popular krypton fluoride (gas) laser. Graduate students could build such a laser for between $10,000 and $40,000—a repeat of the popularity of building high-power carbon dioxide infrared gas lasers a few years back? Krypton fluoride emits in the ultraviolet (2484 angstroms), and, as a gas laser, does not have cooling problems and might therefore be able to generate pulses containing 1 joule and lasting 5×10^{-9} second at a rate of 10 to 20 per second. However, a krypton fluoride laser with 10-joule pulses of 10^{-9} second duration would go back over the $100,000 mark.

A recent development by Richard McCorkle and his colleagues at IBM's Yorktown Heights laboratory could affect the cost issue, at least for x-ray lithography. The IBM researchers have constructed an electron beam–induced plasma source that costs less than $10,000 to build (*Science*, 27 July 1979, p. 401). The source consists of a polyethylene capillary with graphite electrodes. An electric discharge through the capillary vaporizes several layers of the inner wall, producing carbon ions (C^{4+}). A beam of electrons traveling at relativistic speeds through the capillary pinches the plasma, thereby forming a small, bright x-ray source akin to that generated by a focused laser beam. The dominant x-ray emission is in a line centered at 300 electron volts. The duration of the x-ray pulse is rather long, 60×10^{-9} second.

To demonstrate the efficacy of their source, the investigators chose an application that is drawing more and more attention from biologists—contact soft x-ray microscopy. According to Ralph Feder of IBM, work with a plasma source to do x-ray lithography, which is quite similar to contact microscopy but requires more energetic x-ray photons (about 1000 electron volts) and therefore a plasma containing something other than carbon ions, is just getting under way. There is no doubt that one of the world's largest manufacturers of microelectronics circuits would be interested in ways to make them still smaller.

In contact microscopy, the specimen replaces the metal mask used in lithography. In this way, a shadowgraph of the specimen is recorded in a photosensitive polymer. After the polymer is developed and is coated with a thin metal layer, a scanning electron microscope is used to view the shadowgraph. As with lithography, soft x-rays are ideal, especially for the biological structures of most interest, because they are easily absorbed, allowing high-contrast images to be obtained. Moreover, imaging of living organisms is made possible because the soft x-rays emitted by the carbon ions are not absorbed strongly by water. In one early imaging experiment with diatoms (a kind of alga), the researchers achieved a resolution of 300 angstroms, about three times poorer than the best obtained using synchrotron radiation.

All the laboratories working on hot plasma x-ray sources have, in the past, also developed ideas for x-ray lasers. While the applications of plasma sources are in their infancy with much room to grow, one cannot help but wonder if hot plasmas might also be a way-stop on the road to an x-ray laser.

—Arthur L. Robinson