Section 3
TECHNOLOGICAL DEVELOPMENTS

3.A A Facility for Time-Resolved Low-Angle X-Ray Diffraction Studies

In a previous report[1] we described preliminary results on time-resolved x-ray diffraction from a photostimulated biological system, the purple membrane of the *Halobacterium halobium.* Those results were obtained on a partially completed x-ray-diffraction camera system under development in the GDL experimental facility. In this report, we describe numerous improvements to the facility which permit the single-shot recording of structures having periodic scale lengths between 3 Å and 250 Å. We illustrate this with recent examples of membrane diffraction obtained from various systems. The performance of this laser-driven x-ray-diffraction facility is compared with what is available on other systems, and the prospects for further improvements in performance are summarized.

While the production of intense bursts of x-ray radiation is coincidental in direct-drive inertial-fusion target experiments, laser-produced plasmas may also be employed for the express purpose of generating intense x rays. For laser-heating pulses of a nanosecond or less, the hot, high-density plasma formed near the surface of a solid target by a single, focused laser pulse has several important characteristics as an x-ray emitter. The dimensions of the x-ray-emitting region are small, with the lateral extent comparable to the size of the laser focal spot. The x-ray emission, especially for photon energies above one kilovolt, closely follows the laser-pulse time history. Depending on the choice of target,

intense monochromatic line radiation may be produced at many wavelengths between 1.5 Å and 5 Å. Recently, the greatly improved plasma heating characteristics of the frequency-tripled Nd: glass laser radiation from GDL has enabled us to produce such monochromatic line radiation near 3 keV with a conversion efficiency (of laser energy into x rays) of 1 to 2%.[2]

Since the x-ray source is driven by a laser, it may readily be synchronized with a wide variety of external events and thus serve to create a probe beam for the study of kinetic structural phenomena down to atomic-scale resolution. Because the x-ray emission is, roughly speaking, isotropic, the design of a suitable x-ray probe-beam facility presents a challenge in maximizing the collection and transport of x rays. In our application, namely low-angle x-ray diffraction, the net divergence of the beam translates directly into the angular resolution of the system, so our x-ray optical system was constrained to be a focusing system. The point nature of the laser-driven source is very important in that it permits a simple, high-efficiency focusing system to be constructed at grazing angles of reflection. Our design is shown schematically in Fig. 18. We use a toroidal mirror in the form of a truncated ellipsoid to focus the x rays.[3] In this geometry it can be shown that the maximum useful collection solid angle is of the order of θ_c^2, where θ_c is the critical angle for total reflection of x rays. The critical angle depends upon the material properties of the mirror surface as well as the

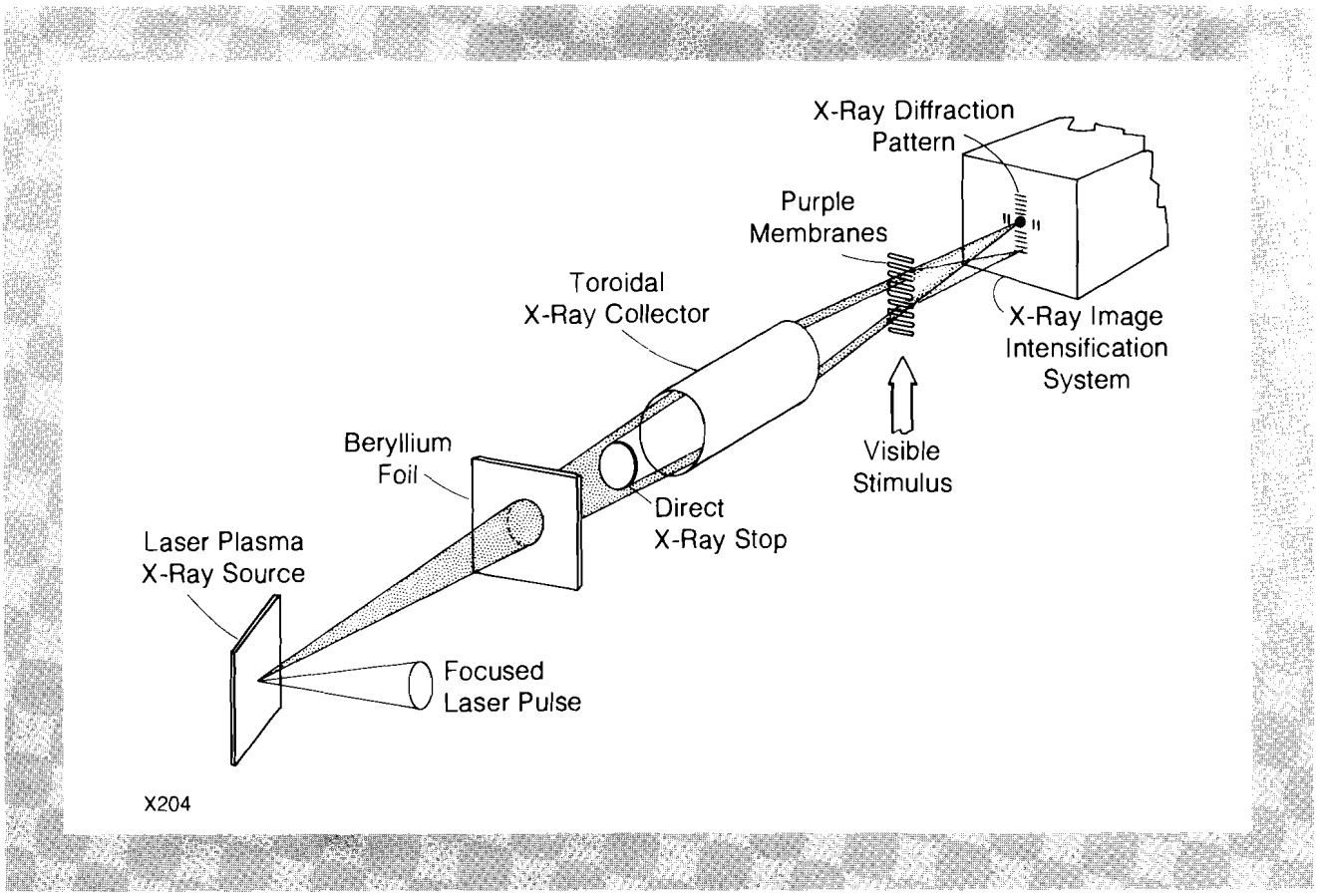

Fig. 18
Conceptual layout of pulsed x-ray-diffraction experiment.

x-ray wavelength. Nickel is a good choice for reflection of x rays in the 2- to 5-Å range and results in a collection solid angle of approximately 2×10^{-4} sr for such a system. Our mirror was fabricated from a CERVIT™ substrate in the optical shop at the University of Rochester and coated with nickel in the LLE target-fabrication facility. It has a focal length of 86 cm.

The specular beam reflected from this mirror has an annular structure which must be taken into account in preparing and aligning specimens in the camera. A space of approximately 20 cm is available in front of focus to position specimens. This space is isolated from the rest of the laser-plasma target chambers by a vacuum window, allowing the specimen environment (temperature, atmosphere, humidity, etc.) to be controlled by the experimenter. On the vacuum side of the window, a narrow annular stop absorbs most of the non-specular x rays scattered from the finite microstructure of the mirror surface. In recent x-ray diffraction experiments, 10^{10} photons of foil-filtered monochromatic x rays at 4.45Å were routinely delivered to specimens on each GDL laser shot.

It is interesting to compare the performance of this low-angle, beamline facility with both conventional laboratory facilities and synchronous electron-storage-ring facilities. In Fig. 19 we plot

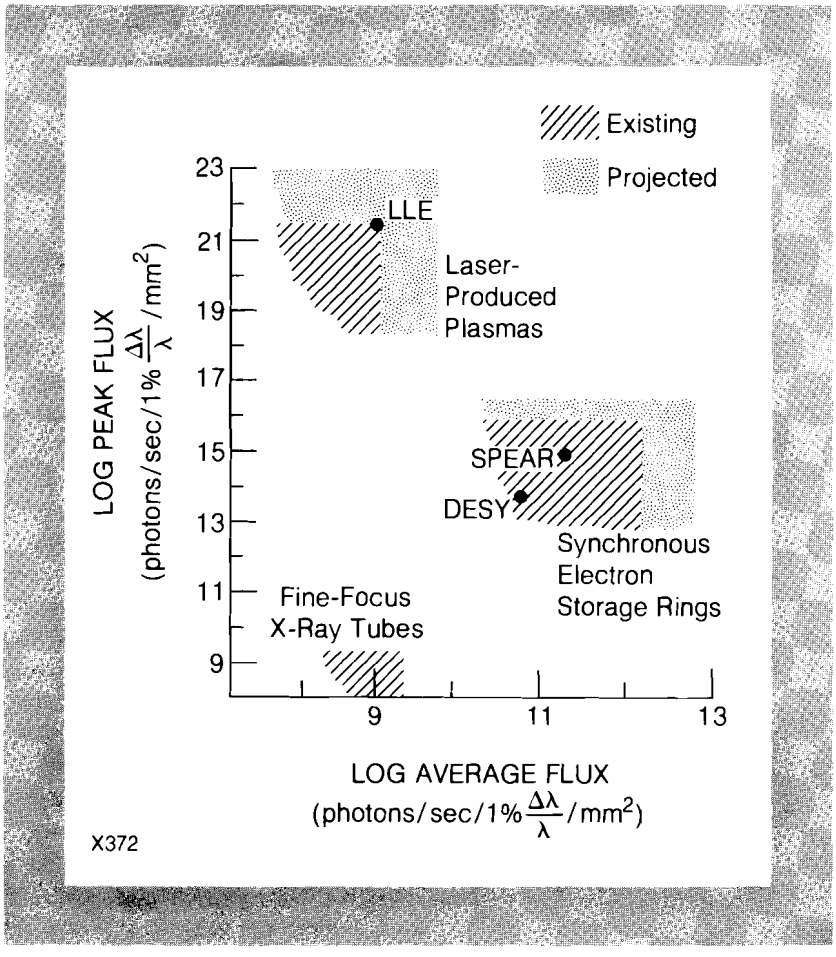

Fig. 19
Peak versus average flux delivery of typical x-ray sources in low-angle diffraction experiments.

the peak and average flux delivery capabilities of these systems. It is clear that the GDL facility can generate diffraction data at a rate equal to the best laboratory facilities while doing so with a potential time resolution many orders of magnitude faster than storage-ring facilities. In order to realize this level of performance, however, great care must be given to the design of the detection system in laser-plasma experiments. Because of the unprecedented delivery rate of x rays in this case, it is impossible to use counter tubes of any currently available design as detectors. Solid-state detectors have many desirable characteristics but are not yet available in sufficiently large-area arrays to be suitable for this application. On the other hand, because of the windows used, TV detectors have no soft-x-ray response. This latter problem is overcome by using a thin scintillator material to convert the x rays to visible light. Although this latter idea is simple in principle, its satisfactory implementation into our system proved very difficult until recently, because of a subtle property of the high numerical aperture of the x-ray focusing optics: the true focal surface of the camera is a sphere tangent to the focus of the camera and to the specimen. The effects of this curvature are not noticed in long-focal-length or low-numerical-aperture systems because they exhibit a large depth of focus.However, in our preliminary experiments on diffraction from purple membrane[1] we were limited to observation at low scattering angles, in part because of serious defocusing near the edges of our flat scintillator plate. Even with a 40-mm-diameter detection field, observation of the highest-angle diffracted orders from purple membrane required a specimen position only 3.5 cm from focus.

Uniform deposition of a thin scintillator onto steeply curved surfaces by commercial vendors has proved to be difficult. It appeared that we could acquire data of satisfactory quality with a focal sphere 5 cm in diameter, and success in such a scintillator deposition was achieved by Thomas Electronics, Inc. In our present camera, the scintillator is deposited on a special fiberoptic plate in which the individual fiber axes are at near-normal incidence to the curved scintillator surface as well as to the flat output surface.

Photons from the scintillator are coupled directly to a VARO 40-mm microchannelplate (MCP) image intensifier set to a gain of 40,000. The MCP output is then lens-coupled to a commercial, refrigerated, digital TV detector (Princeton Applied Research OMA II) operated in a fully two-dimensional mode by means of an LSI 11/23 computer system. The TV detector is operated at −25°C with multiple, high-voltage, slow-scan readout of the image for maximum sensitivity and linear dynamic range.

The performance which may be obtained from our x-ray-diffraction camera is shown in Fig. 20. This is a powder diffraction pattern obtained from a dried pellet of purple membrane with a single shot of 45 J at 0.351 μm from GDL. A sector of the diffraction pattern was circularly averaged in the LSI 11/23 computer to

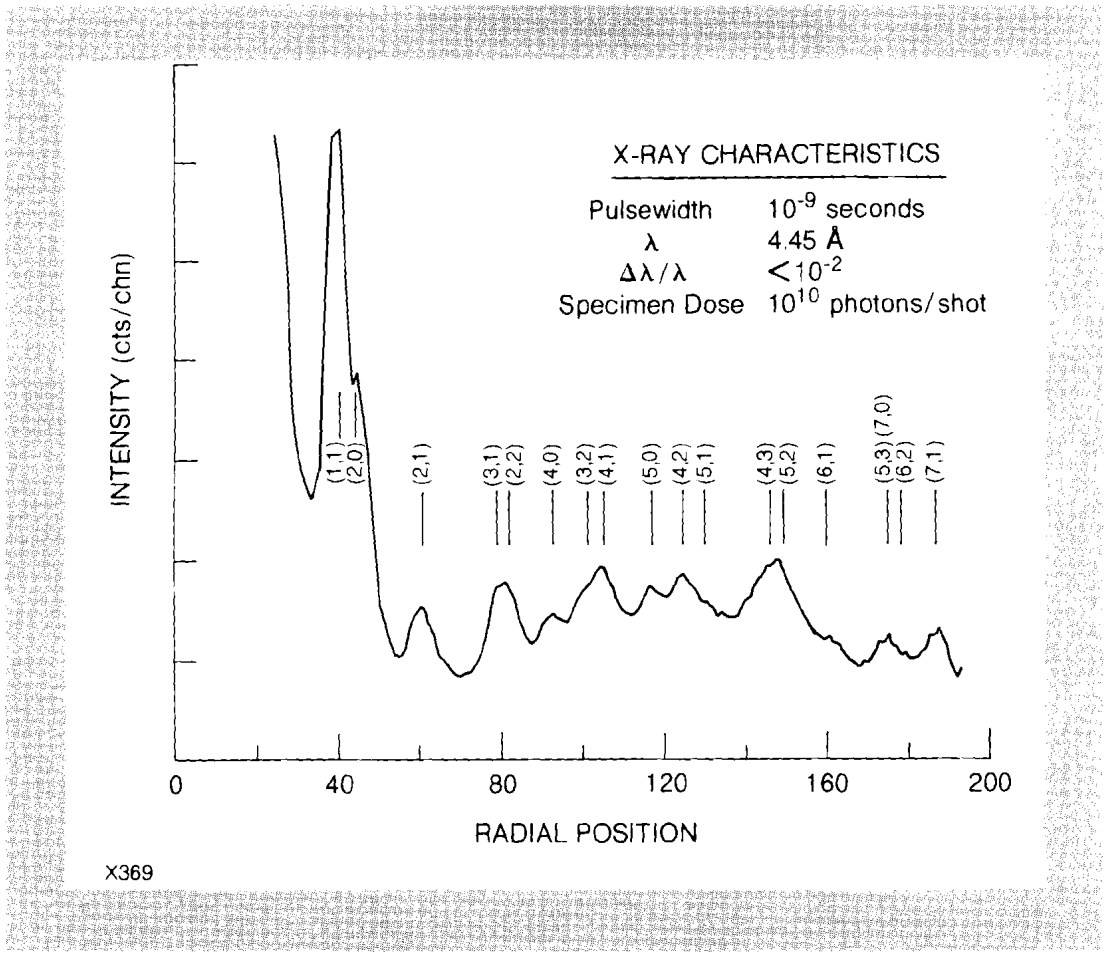

Fig. 20
Azimuthally averaged powder x-ray diffraction pattern from a dried pellet of purple membrane from "Halobacterium halobium." Peaks are shown indexed on a hexagonal, two-dimensional lattice out to 7-Å resolution. A single shot from the frequency-tripled GDL facility was used to acquire this pattern.

produce this display. The specimen consists of an array of membrane fragments which are primarily two-dimensional in crystalline character. The reflections are indexed on a two-dimensional hexagonal lattice after Henderson.[4] The highest-order reflection shown corresponds to 7-Å resolution in the plane of the membrane (and is the highest-order reflection to be reliably phased by any group to date). Experiments to observe structural changes in purple membrane on photostimulation, begun some time ago, have greatly benefited from this improved performance. Significant alterations in the relative intensities of many of the high-order reflections are observed 1 ms after photostimulation of the specimens while little change is noted 200 μs after photostimulation.[5] This timescale corresponds to the time in the purple-membrane photocycle when hydrogen-ion release occurs. The results of further experimentation and analysis on the purple membrane will be presented in a future report.

Other membrane ion-transport experiments are also being conducted, in collaboration with NLUF participants. To date these experiments have concentrated on studies of sample preparation, and handling problems associated with the unique configuration of our camera. A preliminary experiment performed during the quarter recorded a diffraction pattern from a layered

preparation of sarcoplasmic reticulum prepared by J. Kent Blaise and Donnatella Pascolline (University of Pennsylvania). Future reports will give further details of these experiments. For the present report, we note that this specimen is characterized by a rather large basic structural period of the order of 200 Å or so, depending on the relative humidity. A display of a single-shot diffraction pattern recorded on our TV detector from this specimen is given in Fig. 21. Our ability to observe the first-order peak in the diffraction pattern is significant in that kinetic studies of relatively long-period samples such as polymer systems would appear quite feasible with this apparatus.

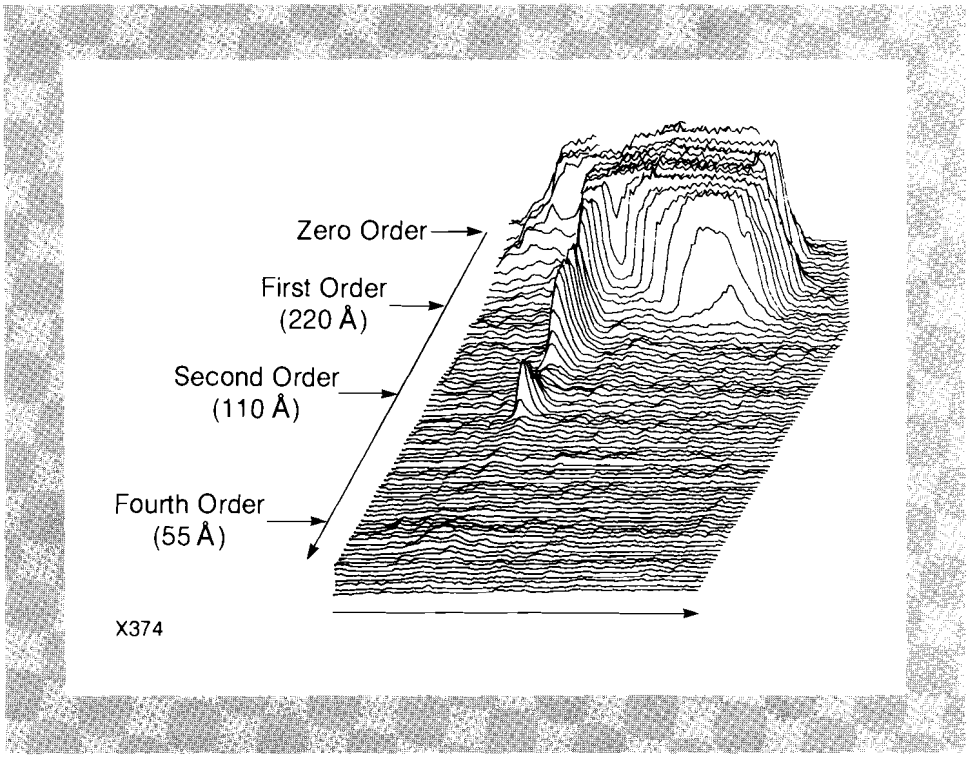

Fig. 21
Two-dimensional display of a single-shot x-ray diffraction pattern from sarcoplasmic reticulum. The first-order peak corresponds to a period of 220 Å.

Single-shot diffraction from a synthetic multilayer, composed of dipalmitoyl lecithin, is shown in Fig. 22. Although the peak of the first-order (55-Å) scattering has saturated the detector, the disorder in the multilayer (mosaic spread) is evident from the arc-like scattering around the peak. The lecithin compound was also prepared by the University of Pennsylvania group as a calibration compound for the sarcoplasmic-reticulum experiment. This pattern also shows the low-angle capabilities of the facility.

The biological systems studied to date, although relatively weak x-ray scatters, are nevertheless ordered systems. It is of interest to some workers to study structural kinetics in poorly ordered systems such as solutions. Many chemical or biochemical reactions such as enzyme-substrate interactions, protein folding, etc. could be studied in detail if the available x-ray fluxes could be increased. In fact, we anticipate such a flux increase in our system toward the end of 1983 with the completion of the active-mirror upgrade and a larger-aperture frequency conversion of the GDL

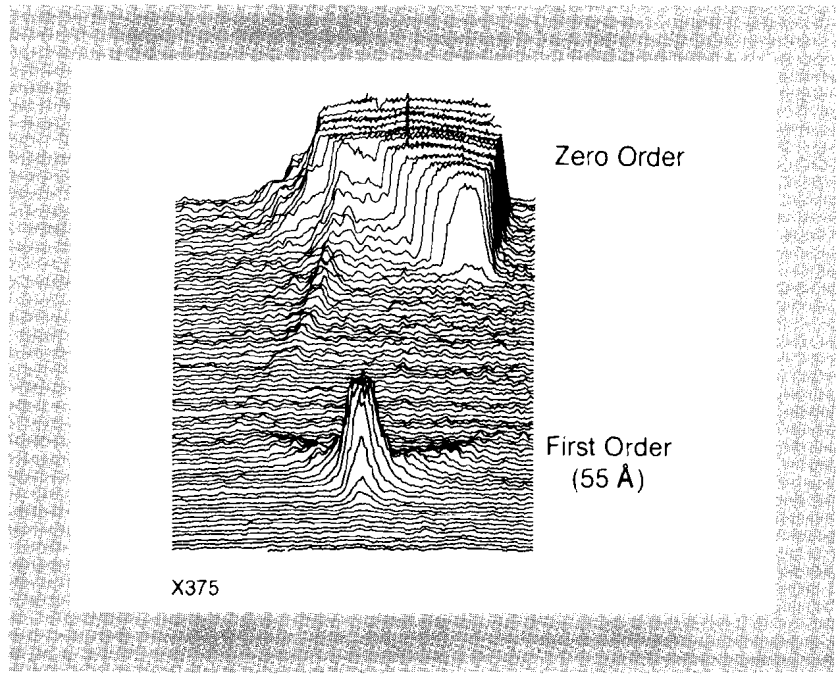

Fig. 22
Two-dimensional display of a single-shot x-ray diffraction pattern from dipalmitoyl lecithin. The first-order peak has saturated the TV detector but the intense signal level permits direct observation of the mosaic spread in the sample from the arc-like scattering surrounding the peak.

facility. When combined with expected improvements in x-ray yields from highly chlorinated target materials, we expect to deliver up to 10^{11} x-ray photons per shot to specimens in our camera.

Summary

The past two decades have seen extensive development of high-average-power x-ray sources with great improvements in conventional structural measurements, such as crystallography, and the introduction of entirely new techniques, such as extended x-ray-absorption fine structure (EXAFS). The introduction of the laser-driven x-ray source as a structural analytic tool will have an impact on a broad range of scientific and technological problems by greatly extending our capabilities for time-resolved measurements of bulk structures.

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