High-Resolution Imaging with Soft X-rays

The first fruit of the emerging field of soft x-ray optics is likely to be a scanning x-ray microscope for biologists

"In x-ray optics, there are no new ideas, only new technologies to exploit," says Natale Ceglio of Lawrence Livermore National Laboratory. Researchers have always had ample notions of how to take advantage of the short wavelengths of x-rays as compared to visible light, but they did not have the ability to carry out their ideas. The lenses, mirrors, and gratings that work so well in the visible cannot handle x-rays.

Yet, in the last decade, astronomers using x-ray telescopes orbiting in satellites, physicists studying the properties of plasmas for fusion energy, and researchers exploiting the intense, tunable x-ray beams from synchrotron radiation sources have all been driven to develop ways to collect, disperse, focus, and image x-rays with wavelengths from 100 angstroms to less than 1 angstrom. Two conferences, this summer and fall, on soft x-rays (10 angstroms or longer) show how the common interests of this diverse group and the increasing ability of microfabrication practitioners to make structures with dimensions comparable to the wavelengths of x-rays have combined to launch the discipline of soft x-ray optics.*

Some observers say that the optical capabilities becoming available in the soft x-ray region will also come later on for the shorter wavelength, hard x-rays. In the meantime, soft x-rays are exciting enough. Biologists, for example, find this wavelength range useful because soft x-rays can penetrate thick samples consisting largely of water but are absorbed by carbon-containing molecules. Thus, high-contrast images of wet or even live specimens is possible. In fact, among the many applications of soft x-ray optics, the one currently drawing the most attention is the development of an x-ray microscope. The best instrument at present has a resolution of about 500 angstroms, but 100 angstroms is only a year or two away. The wished-for resolution previously achievable only with electron microscopes, but without the need to thin, dry, and stain specimens, is about to be fulfilled.

Two kinds of devices appear especially promising for high-resolution soft x-ray optical systems: Fresnel zone plates and multilayer metallic mirrors. In both cases, microstructure fabrication processes are essential for making devices that work with soft x-rays.

Fresnel zone plates are circular devices consisting of concentric rings and having some resemblance to a diffraction grating. The rings are alternately x-ray absorbing and transmitting. The area of each ring is the same, so the outer rings get progressively narrower. Zone plates act like lenses: a parallel light beam is focused to a point, and so on. A lens made entirely of transmitting materials, and a lens thick enough to have a focusing effect would absorb all the x-rays first. Working on a diffraction principle, the zone plate does not need to be so thick.

The resolution of an image formed with zone plates is determined by the width of the outermost ring. In his talk at the Topical Conference on Low Energy X-Ray Diagnostics in Monterey last June, Ceglio concluded that for a microscope using 50-angstrom x-rays to achieve a resolution of 200 angstroms, a zone plate with gold absorbing rings would have to have 100 rings, and the width of the outermost ring would have to be 200 angstroms or less. For comparison, the most advanced commercially available integrated circuits have as their smallest structures metallic conductors that are more than 100 times as wide, although researchers can do considerably better in the laboratory. Ceglio pointed out that Michael Hatzakis of IBM's Yorktown Heights laboratory several years ago used a computer-controlled scanning electron microscope to write a zone plate pattern with a minimum zone width of 700 angstroms.

The idea of using zone plates as the optical elements in an x-ray microscope dates back to at least the 1961 experiments of Albert Baez of the Smithsonian Astrophysical Observatory. Now a number of projects are under way that rely on the intense x-ray beams available from synchrotron light sources as the illuminating medium. The most advanced of these is the one at the University of Göttingen by Günter Schmahl, Dietbert Rudolph, and Bastian Niemann. At the Society of Photo-Optical Instrumentation Engineers (SPIE) conference on High Resolution Soft X-Ray Optics, held at Brookhaven National Laboratory in November, Rudolph showed photomicrographs taken with the newest version of the x-ray microscope, which achieves a resolution of about 500 angstroms. Niemann discussed the group's plans for a 100-angstrom-resolution instrument that is now under development and may be operating in the next year or two.

Rudolph says the group began its research in 1967 with the invention of the holographic grating. Two intersecting coherent beams of light from the same laser produce an interference pattern of bright and dark lines whose spacing is related to the angle between the beams. A photoresist (polymer sensitive to visible or ultraviolet light) placed at the interference pattern location, exposed, and developed then serves as the template for grating manufacture. A year later, the Göttingen group extended the holographic technique to the manufacture of zone plates. Two laser beams passing through a complex (aplanatic) system of six lenses were used to make the required pattern of concentric circles of increasing radius and decreasing width.

In 1975, the investigators went to the German Electron Synchrotron (DESY) laboratory in Hamburg to begin prototype imaging studies with the bright light from the electron storage ring DORIS. Schmahl, Rudolph, and Niemann later accepted an offer to work at a much lower energy electron storage ring, ACO, at the Laboratory for the Utilizational of Electromagnetic Radiation (LURE) in Orsay, near Paris, whose spectrum of synchrotron radiation is more suitable for soft x-ray experiments.

The first microscope constructed by the group consisted of a long, evacuated tube. Within the tube, a diffraction grating at grazing incidence selected a nar-

Low Energy X-Ray Diagnostics 1981, D. T. Attwood and B. L. Henke, Eds. (American Institute of Physics, New York, 1981); *SPIE Proceedings*, vol. 316, E. Spiller, Ed. (Society of Photo-Optical Instrumentation Engineers, Bellingham, Wash., to be published in 1982).

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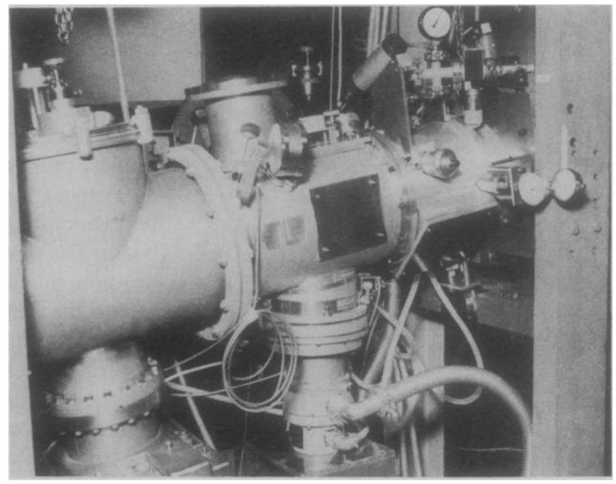
row band of x-ray wavelengths from the continuous distribution emitted by ACO. A condenser zone plate 4.5 millimeters in diameter with 15,000 rings focused the resulting monochromatic soft x-ray beam onto the specimen, which was in a special chamber that permitted examination of wet, live specimens. A second, high-resolution micro-zone plate only 3 micrometers in diameter with 625 rings imaged the transmitted beam from the specimen onto a detector. With 44-angstrom x-rays, this apparatus achieved a resolution of about 1200 angstroms in such specimens as diatoms and kidney and liver cells.



X-ray microscope

Synchrotron radiation enters from the left. The condenser zone plate, the specimen chamber, the micro-zone plate, and the detector positions can all be adjusted from outside the evacuated microscope during an experiment. [Source: D. Rudolph, University of Göttingen]

An improved instrument eliminated the grazing incidence diffraction grating in the interest of getting more of the incident light beam onto the specimen; the grating transmitted only about 10 percent of the incoming x-rays. It was possible to do without the grating because zone plates are highly chromatic; that is, they focus light of different wavelengths at different distances from the plate. Thus, a zone plate together with a pinhole to block the unfocused wavelengths acts as a monochromator to select a narrow band for transmission. The improved instrument also incorporated a much smaller imaging micro-zone plate with 100 rings and an outermost ring width of 580 angstroms, as compared to the 1200 angstroms in the earlier version of the microscope. At the SPIE conference, Rudolph showed 500-angstrom resolution photomicrographs of diatoms taken with this microscope. Exposures were for 20 seconds or less with x-ray radiation of 45 angstroms.

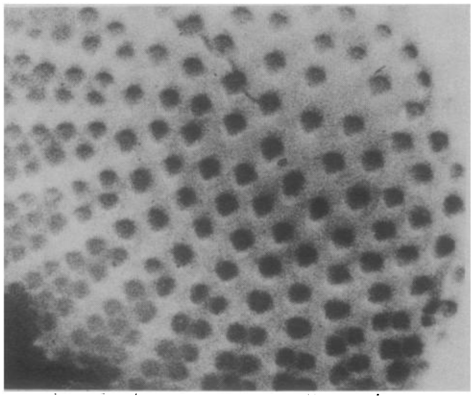
Niemann followed Rudolph's presentation with a description of plans for a scanning x-ray microscope with 100-angstrom resolution. Although an image must be built up point by point and therefore takes a longer time (Niemann estimated that a picture of 200 by 200 points would take 40 seconds) than direct imaging, the radiation damage to the specimen is reduced 100-fold. This is partly because the position of the micro-zone plate and the specimen are reversed, so that only radiation that will actually be used in constructing the image strikes the sample. Just as important, a more efficient detector can be used, which detects every photon passing through the specimen. In conjunction with the ability to select a wavelength from the broad synchrotron spectrum that is absorbed strongly by only one element, scanning also allows mapping at high resolution the distribution of atomic species in wet or dry specimens.

A crucial feature of the scanning instrument will be the micro-zone plate. Niemann described a process whereby a rotating wire would be alternately coated with thin layers of x-ray absorbing (gold) and transmitting (boron) materials. The wire could then be sliced transversely, and the slice removed would be the zone plate. Thicknesses of coatings can be controlled to within a few angstroms, so a zone plate with an outermost ring width of 100 angstroms is quite feasible. Niemann also outlined the features of a movable stage that could control the position of the specimen to 100 angstroms or better. This is necessary because the x-ray beam cannot be scanned the way an electron beam can by use of electric or magnetic fields.

The Göttingen group hopes to have this microscope working in 1982. It will be set up in Berlin, where a new electron storage ring, BESSY, is nearing completion. BESSY will provide a much more intense soft x-ray beam and one that is much smaller in diameter than those now available and hence will be very well suited for scanning microscopy. Rudolph says that the group's strategy is to demonstrate a 100-angstrom resolution on test samples. The idea is that when biologists see that the instrument really works as advertised, collaborations can be established that will address serious problems that need images of this resolution for progress to be made. The three-dimensional arrangement of specialized regions within cell membranes is one example that has been suggested.

In the United States, Janos Kirz and Harvey Rarback at the State University of New York at Stony Brook are working on a scanning x-ray microscope to be in place in the next 2 years at the National Synchrotron Light Source at Brookhaven National Laboratory, where there is an ultraviolet-soft x-ray electron storage ring of performance similar to BESSY's. The Stony Brook researchers will use a grating and a single zone plate of a special type. A large number of the central zones are opaque as if there were a huge bull's eye filling half the zone plate (apodized zone plate). This configuration is required to produce a high signal-to-noise ratio when only a single zone plate is used. The first microscope will have a modest resolution of about 1000 angstroms.

The second new type of soft x-ray optical device is the multilayer mirror. Because their index of refraction does not change much on going from air to a solid material, x-rays striking a surface perpendicularly (normal incidence) are only weakly reflected by polished metal surfaces. Reflection is most effective at grazing angles of incidence, and the x-ray optical systems in satellite x-ray telescopes and in the x-ray beam lines at synchrotron radiation centers have relied on grazing-incidence mirrors to guide and focus radiation. The idea behind multilayer mirrors is to create several (a hundred or more in some cases) reflecting interfaces between materials that absorb x-rays and those that transmit x-rays, all in a distance shorter than that in which x-rays are completely absorbed. If the layers have the correct thickness, then the small amount of reflected x-ray light at each interface can add constructively with the others. The overall effect is a reflectivity of 10 to 20 percent for x-rays at normal incidence as compared to less than 0.01 percent for a single layer. A multilayer coating on a spherical mirror could make a very effective x-ray focusing device, for example. In general, the constructive interference criterion is satisfied only for a narrow band of x-ray wavelengths, so multilayer mirrors also function as a filter or monochromator when a broad band of radiation strikes them. Two groups, one at Stanford University and one at the IBM Yorktown Heights laboratory, have con-



X-ray micrograph of a diatom

The Göttingen group used 45-angstrom synchrotron radiation from the ACO storage ring at Orsay to produce this image with magnification ×160 and resolution 500 angstroms with a 20-second exposure.

centrated on the synthesis of multilayer mirrors, and both reported on their progress at the Monterey meeting.

At Stanford, Troy Barbee and Douglas Keith use a sputtering technique to make their multilayers. Barbee reported that individual layers as thin as 7 angstroms were made in their apparatus. Materials sputtered included tungsten, niobium, molybdenum, titanium, vanadium, and silicon. These materials are x-ray absorbers. Carbon was used as an x-ray transmitting material in most mirrors. But work with boron and magnesium is also in progress. The use of materials other than carbon means that the important range of soft x-ray wavelengths for biological applications, where carbon becomes an absorber, can be used.

The Stanford investigators use a so-called enhanced sputtering source (magnetron) which has two advantages. The first is that the deposition rate of this notoriously slow process is increased, although it still takes about an hour to make a multilayer mirror. The second is that the plasma of ionized gas, which dislodges atoms from the surface of a target made of the material to be deposited on a substrate some distance away, is localized near the sputtering target. There is thus no radiation damage to the mirror that is gradually building up on the substrate. There is a separate magnetron source for each material to be sputtered, and the substrate is on a movable stage that rotates to a position appropriate for each source.

In one series of measurements with tungsten-carbon mirrors of varying layer thickness, Barbee reported normal incidence reflectivities as high as 18 percent. In general, the reflectivity decreased with decreasing spacings between the layers, especially below a layer thickness of 15 angstroms. Barbee attributed the falloff of the reflectivity to a slight roughness in the interfaces between the layers of about 3 angstroms.

The IBM group, headed by Eberhard Spiller, makes its multilayer mirrors by an evaporation process. The material to be evaporated is heated by an electron beam. The thickness of the layer building up on a substrate is monitored by measuring the reflectance of x-rays from a small x-ray tube in the vacuum system. Separate electron beam heated sources are used for the different materials in the layers, and shutters protect the fixed substrate from being contaminated by a source not in use.

Like the Stanford group, Spiller and his colleagues use silicon substrates because they are generally the smoothest available, but they have also experimented with glass. They have tended toward the use of metal alloys, such as gold-palladium and rhenium-tungsten, as the absorbing layer material and carbon as the transmitting layer. The highest reflectivity was about the same as that at Stanford and was obtained with rhenium-tungsten mirrors. The IBM group also directly measured the surface roughness of the top layer of various layers as they grew and found values of a few angstroms.

At the Brookhaven conference, Spiller summarized the progress of his group toward the construction of a soft x-ray microscope built around spherical mirrors coated with multilayers rather than around zone plates. An instrument has been assembled, but the researchers cannot test it until the synchrotron radiation source at Brookhaven is fully operational. Conventional sources are not intense enough for scanning microscopes. Moreover, the wavelengths for the peak reflectivities of the first multilayer mirrors, which are made of rhenium-tungsten and carbon with thickness designed to reflect 68-angstrom x-rays, did not quite match the wavelengths of conventional sources that emit at only a few wavelengths.

The instrument is called a Schwarzschild microscope. It consists of two spherical mirrors, one having a radius roughly twice that of the other. X-rays enter the microscope through a hole on the axis of the larger diameter mirror and are reflected from the convex surface of the smaller diameter mirror back toward the concave surface of the larger mirror, where a second reflection focuses the light onto a specimen. Spiller reported that he expects the instrument to have a resolution of about 500 angstroms. The quality of the surface of the spherical mirrors is the main limitation, and Spiller thinks that a reduction of variations (figuring errors) in the spherical surfaces from the present state-of-the-art 50 angstroms to a few angstroms is achievable and will permit a Schwarzschild x-ray microscope to have a 200-angstrom resolution or better. He cited a claim by the Perkin-Elmer Corporation that it will be able to test spherical mirrors to within 6 angstroms by next year as one example of the coming capability.

From this account, it would seem that Fresnel zone plates and multilayer mirrors are somewhat competitive approaches toward an x-ray microscope. Many researchers, however, do not share Spiller's optimism about the likelihood of making more perfect mirror surfaces. The view of these observers is that multilayer mirrors will more likely find application in places where a very high resolution is not required but the ability to make large-surface-area optical devices is needed. Large x-ray telescopes with 0.1 arc second resolution (30 times better than the Einstein satellite) would be one example of this kind of application. The large light-collecting surface of mirrors is needed to gather in as many x-rays as possible.

Researchers working with soft x-rays are clearly delighted by the progress of the last few years. One remaining problem is that the field is still fragmented, with only a little communication between those working in different disciplines but having a common x-ray interest. The recent conferences have helped to bring this community together. But some researchers think that further rapid progress may require a less haphazard approach to x-ray optics. One way to focus the effort would be to establish a national or regional institute devoted to this goal. In the United States, Ceglio and David Attwood of Livermore are spearheading an effort to establish such an institute. The project is at such an early stage, however, that even such a detail as where it would be located is still undecided. In Europe, institutes or national centers are in the embryonic stage in France, West Germany, and the United Kingdom. For example, Schmahl at Göttingen is beating the bushes to find support for an institute there. In all cases, the biological applications of the x-ray microscope may be the key. Attwood and Schmahl say that they have been advised that an important part of selling their projects will be finding biologists who are excited about the prospects of imaging with x-rays and will be willing to campaign for an institute with the facilities that are required for this kind of work.—ARTHUR L. ROBINSON