Section 4
DEVELOPMENTS IN SUBPICOSECOND RESEARCH

4.A Picosecond Electron Diffraction

Laser-induced structural kinetics in the picosecond time domain are of current interest in the field of solid state surface physics and semiconductor annealing. In the past, time-dependent structural kinetics have gone unmonitored for lack of a suitable picosecond x-ray or electron probe in synchronism with the laser pulse. Recently, subnanosecond x-ray bursts produced by a laser plasma have been successfully used by R. Frankel and J. Forsyth[1] to generate low angle x-ray diffraction patterns of biological samples. On a slightly longer time scale using synchrotron radiation, nanosecond studies of the kinetics of annealing have been made by B. C. Larson[2] and co-workers. In this article, we report on a technique which makes possible laser-induced structural kinetic studies in the picosecond range using a burst of electrons from the front end of the streak camera.

With the exception of picosecond photoelectron switching recently demonstrated,[3] streak camera tubes have been used exclusively as a fast optical or x-ray diagnostic tool. However, some of the most beautiful features of the image converter tube used in the streak camera have been only partially exploited with this application. The image converter tube produces a temporal and spatial monoenergetic photoelectron replica of an optical pulse. This replica is ultimately limited by the temporal and spatial resolution of the particular streak tube employed. With a relatively low input signal to minimize space charge broadening, the temporal and spatial resolution can be subpicosecond[4] and of the order of 100 μm respectively. Other features worth noting about this photo-

electron replica are that it is synchronized to within a picosecond with the original optical pulse[5] and can be operated at a high repetition rate.[6] These properties make the front end of the streak camera tube particularly well suited for picosecond electron diffraction.

Unlike x-rays which are more appropriate for examining bulk properties of a material, 20 keV electrons penetrate only a few hundred angstroms or less and thus investigate effects on or near the surface of a specimen. As shown in Fig. 19, our experimental layout is comprised of a demountable Photochron II streak camera tube with the deflection plates removed. An aluminum photocathode is used to allow the tube to be opened to air for insertion of the specimen under study. The specimen is located in the drift region of the streak tube 1 cm from the anode and 29 cm from the phosphor screen located downstream. The electron energy is 20 keV and corresponds to a wavelength of 0.86 Å. The specimen thickness is chosen according to the mass thickness parameter $\mu_S = \mu \cdot d$ where μ is the density of the specimen and d is its thickness. The mass thickness parameter must be of the order of $3 mg/cm^2$[7] for a 20 keV electron to experience a single elastic collision.

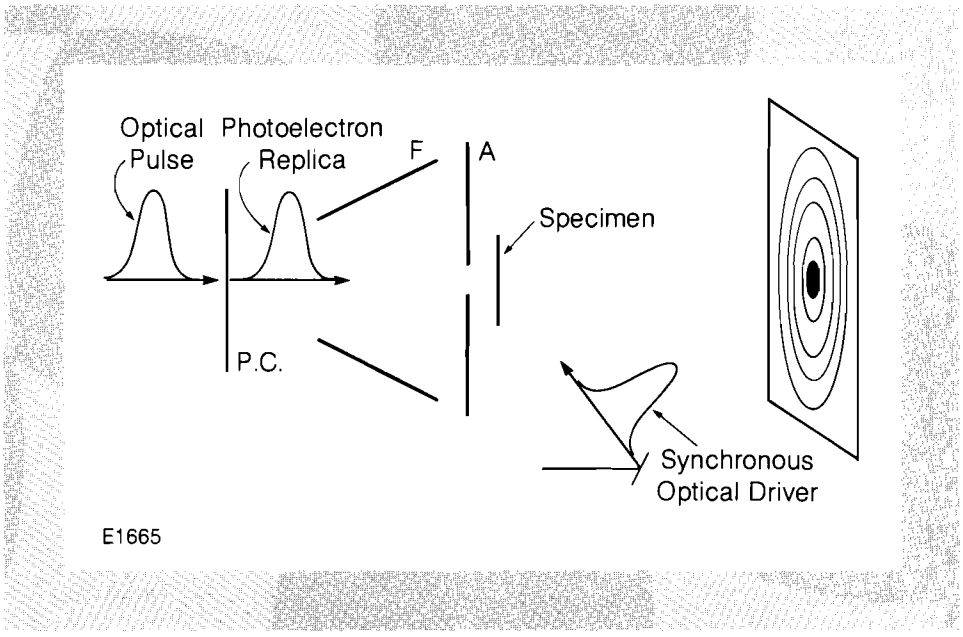

Fig. 19
Experimental layout for generating a picosecond electron diffraction pattern. P.C. is the Al photocathode set at 20 kV, F is the focus cone, A is the anode set at ground potential. The system is evacuated to 10^{-6} mm Hg using an oil diffusion pump.

The fourth harmonic from an active-passive mode-locked Nd:YAG laser is used to overcome the 4.1 eV work function of the aluminum photocathode. At this wavelength (266nm) the quantum efficiency for Al is approximately 10^{-5}. The optical energy on the Al photocathode of the streak camera is approximately a few microjoules. The electron pulse width has been measured by using the camera on the normal streak mode, and is found to be 100 psec. This value departs significantly from the 15 psec pulse width expected. The pulse broadening is due to the space charge effect caused by the relatively high electron flux required to photograph the pattern with our present system. The spot size at the specimen is 3 mm, although a probing cross-section of a few hundred microns in diameter has been used with only a slight reduction in brightness. An image intensifier with a gain of about 10^4 is used to amplify the pattern appearing on the screen.

Figure 20 shows the transmission electron diffraction pattern obtained with a single 100 psec electron pulse passing through a 150 Å thick aluminum specimen. Four distinct rings appear in this pattern. Their radii, after correction for the pincushion distortion of the image intensifier, are in the ratio 1:1.5:1.6:2, and indicate a face-centered cubic type crystal.[8] For each ring the lattice constant a_0 can be found using the Bragg relation for small angles,[8]

$$a_0 = \frac{2\lambda\sqrt{k^2 + l^2 + m^2}}{r} \cdot L \tag{5}$$

where r is the ring radius, L is the distance between the specimen and the phosphor plane, k, l and m are the Miller indices, and λ is the wavelength associated with the electron. For the four rings representing the diffracting planes 111, 200, 220, and 311 (listed radially outwards) we find the lattice constant $a_0 = 4.03 \pm .08$ Å, in close agreement with the known lattice constant for aluminum of 4.05 Å.[8]

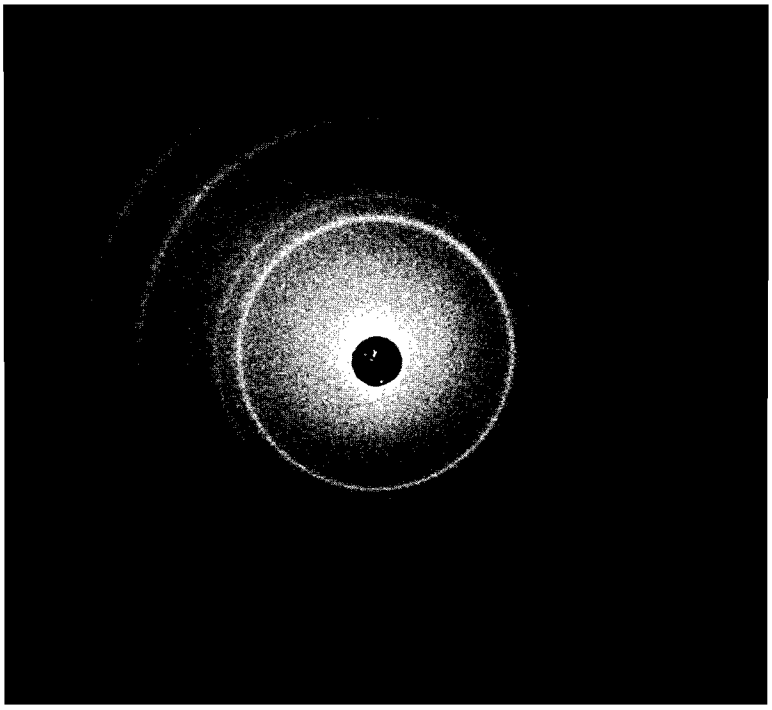

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Fig. 20
Transmission electron diffraction pattern of aluminum from 100 psec exposure of 20 keV electrons. The specimen is 150 Å thick.

The simple experiment described above demonstrates how a short, synchronized electron burst generated by laser radiation impinging on the front end of a streak camera can be used to create an electron diffraction pattern from a thin specimen. Although this pattern is from an electron beam whose pulse width is 100 psec, we expect that this technique can generate electron pulse widths a few picoseconds in duration if presently used electron fluxes are reduced. It should be possible to decrease the electron flux level by a factor of over 100 by using a larger gain image intensifier directly contacting the recording film.

Part of our future work in time-resolved electron diffraction will be directed toward the deflection capabilities of streak camera tubes. By

operating the streak tube in saturation mode so that the space charge significantly broadens the pulse in time, it has been shown that the electron pulse generated in such operation broadens in time about the center, with a width dictated by the current density at the photocathode.[9,10] This temporal broadening can occur with a negligible increase in spot size. The elongated electron burst could be used to probe a specimen over the entire time span of its structural change. The scattered electrons would pass through a thin slit located between the specimen and the deflection plates. The slit would, in the case of the ring pattern, pass small segments of each ring and the deflection plates (with a streak direction perpendicular to the slit length) would streak the time evolution of the changing pattern. In this mode of operation the time resolution is no longer dictated by the electron burst width, but rather by the temporal resolution of the streak region in the streak tube which is at most a few picoseconds.[10] Picosecond synchronization between the laser stimulus and activation of the deflection plates would allow signal recovery techniques to be used.[5]

A further extension of the deflection mode of operation would be to replace the front end of the streak tube with a gated thermionic e-beam, thus removing the need for a photocathode. It should be possible, with an ultra-high vacuum system, to use these techniques to create a picosecond diffraction pattern in the reflection mode. This would permit investigation of surface effects on any thin or thick material.

Presently, an experiment is being carried out using the short electron pulse in transmission mode to investigate the kinetics of a material under laser annealing conditions. We believe that these techniques will make possible the investigation of laser-induced phase transitions in a time domain presently of great interest.

REFERENCES

1. R. Frankel and J. Forsyth, *Science* **204**, 622 (1979).
2. B. C. Larson, Material Research Society, Annual Meeting Boston Paper AI.2. (1981).
3. S. Williamson and G. Mourou, CLEO '81, Washington, D.C. Paper WR5 (1981).
4. D. J. Bradley and W. Sibbett, *Appl. Phys. Lett.* **27**, 382 (1975).
5. G. Mourou and W. Knox, *Appl. Phys. Lett.* **36**, 623 (1980).
6. M. C. Adams, D. J. Bradley and W. Sibbett, *Picosecond Phenomena,* edited by C. V. Shank, E. P. Ippen and S. L. Shapiro (Springer-Verlag., 1978), p. 108.
7. P. Grivet, *Electron Optics* (Pergamon Press, 1972), p. 638.
8. H. P. Klug and L. E. Alexander, *X-Ray Diffraction Procedures* (J. Wiley & Sons, 1967), pp. 319-389.
9. W. Knox (private communication).
10. H. Niu and W. Sibbett, *Rev. Sci. Instrum.* **52**, 1830 (1981).