

2.D Picosecond-Reflection High-Energy Electron Diffraction

Pulsed-laser processing of semiconductors and metals is a rapidly growing field of study. Both the properties of the produced material (which could be unattainable by other means) and the fundamental processes are of great interest. Time-resolved observation of phenomena accompanying pulsed-laser processing has been accomplished by several different techniques. These include optical probing (both reflection and transmission),¹⁻⁵ photoconductivity measurement,⁶ x-ray diffraction,⁷ Raman scattering,⁸ and time-of-flight mass spectrometry.⁹ None of these techniques provides a surface structural probe that can be limited to the first few monolayers. Although second-harmonic generation could provide a surface structural probe,¹⁰ its interpretation is ambiguous.¹¹

Indeed, it is the properties of these initial atomic layers that are of prime importance in laser processing. In addition to its significance in device fabrication, the study of surface-phase transitions and reactions is a rapidly maturing science from both a theoretical and an experimental point of view. The advancement of such knowledge, however, requires the development of a direct time-resolved surface structural probe capable of monitoring the complex processes of surface-phase transitions and reactions. We report here on the demonstration of reflection high-energy electron diffraction (RHEED) with photogenerated picosecond electron pulses. Due to the synchronization of the electron pulses with the laser source, such a technique provides a picosecond time-resolved surface structural probe for material being irradiated with a laser.

Electron diffraction (both high energy and low energy) provides a "natural" surface probe that is well developed and has been used for decades in surface studies. Construction of a picosecond-transmission, electron-diffraction apparatus was accomplished a few years ago.¹² This technique was applied to study the dynamics of laser melting of freestanding thin-aluminum films.¹³ Subsequently, a nanosecond, low-energy electron-diffraction apparatus was developed and used in surface temperature measurement during laser annealing of germanium.¹⁴ In such a setup, due to the low electron energy, space-charge effects severely limited the ability to observe the diffraction pattern. It was estimated that on the average only one electron per shot was reflected from the surface.¹⁴ Thus, averaging over a large number of shots was required to obtain meaningful data. This limitation precluded the use of this technique to study nonrecurring surface structural phenomena. In contrast, in picosecond RHEED, due to the much less severe space-charge limitations, we have demonstrated that excellent quality surface-diffraction patterns could be obtained with a total exposure time of the order of 100 ps.

The picosecond electron-diffraction system, shown in Fig. 32.18, was a modification of the previously described system.^{12,13} The photocathode was $\sim 250\text{-\AA}$ gold film deposited on a sapphire window

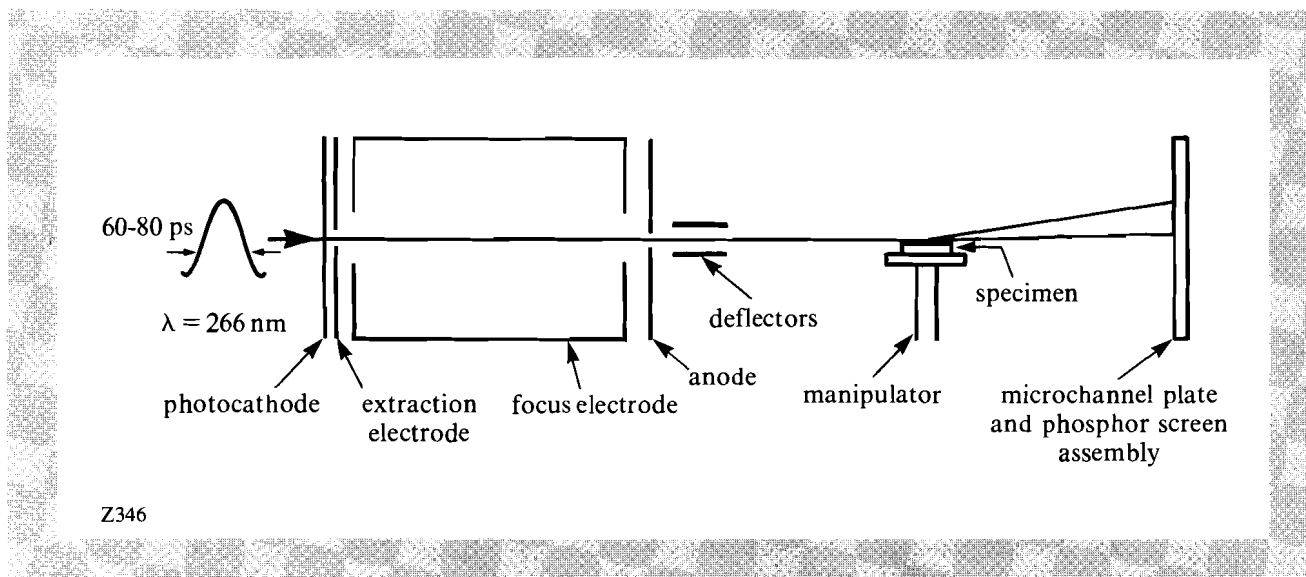


Fig. 32.18

A schematic diagram of picosecond time-resolved-reflection, high-energy electron-diffraction apparatus.

by conventional evaporation techniques. Such a photocathode was rugged enough to withstand months of operation with repeated exposure to the atmosphere. A circular aperture of 0.25-mm diameter located 1 mm from the photocathode was used as an extraction electrode, followed by a focus electrode and an anode that was located 140 mm from the photocathode. The specimen was mounted on a manipulator providing linear motion, rotation, and tilt. The diffraction pattern was amplified by a microchannel plate and observed using a P-47 phosphor screen deposited on a fiber optics faceplate, providing near-zero distortion of the diffraction pattern. The system was typically operated at a pressure of $\sim 2 \times 10^{-6}$ Torr.

The photocathode was activated by UV light from a frequency-quadrupled Nd:YAG regenerative amplifier (an injection-locked multipass amplifier¹⁵). This system provided 0.4-mJ pulses at 1.06 μm and a pulse width of 120 ps to 160 ps. The repetition rate of the regenerative amplifier could be varied from a single shot to 1.5 kHz with shot-to-shot stability better than 5%. Only a very small fraction of the output of the regenerative amplifier was frequency quadrupled ($< 10^{-3}$); thus, the 1.06- μm fundamental wavelength contained almost all the energy and could be used for sample heating. The frequency-quadrupled Nd:YAG ($\lambda = 0.266 \mu\text{m}$) was incident normal to the photocathode and had a ~ 0.1 -mm spot-size diameter on it. The electron replica of the UV pulse, generated at the photocathode, was swept by the extraction aperture and further accelerated and focused in the focus electrode to a focal spot of ~ 0.8 mm on the phosphor screen. These electrons were incident in a grazing angle on the surface of the specimen and Bragg diffracted, giving a RHEED pattern.

Figure 32.19 shows a RHEED pattern of a cleaved surface of sodium chloride crystal obtained using a single-electron pulse and after averaging 50 pulses. The electron energy was 25 keV; a field of

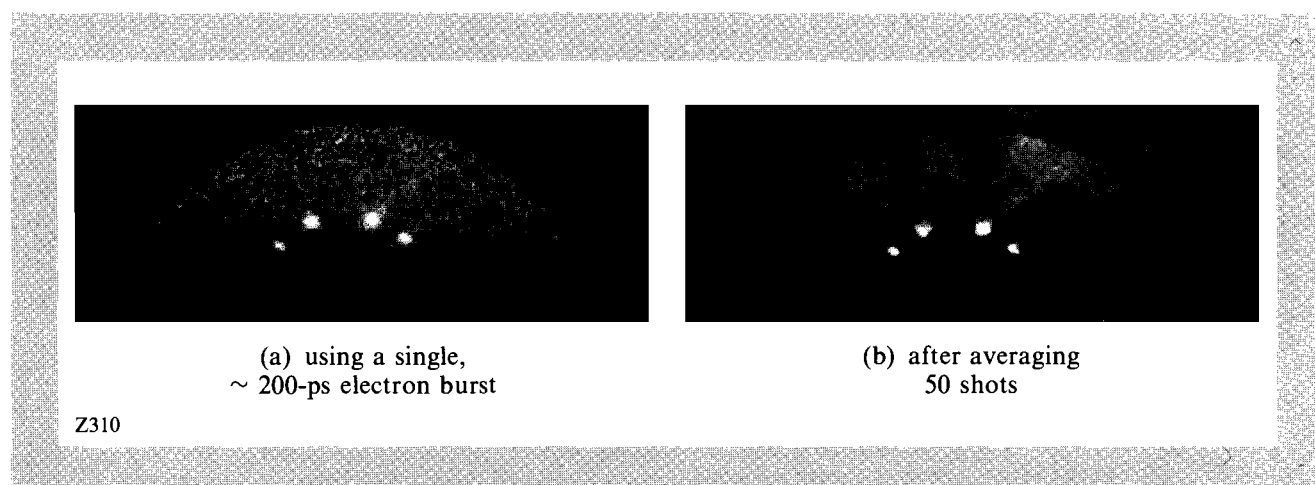


Fig. 32.19

Reflection high-energy electron-diffraction patterns from a cleaved surface of a sodium chloride crystal: (a) using a single, ~ 200 -ps electron pulse; (b) after averaging 50 pulses. The microchannel plate gain was reduced for (b).

23 kV/cm was applied between the photocathode and the extraction electrode. Although the number of electrons per pulse, measured by a Faraday cup and a picoammeter, was found to be directly proportional to the UV intensity on the photocathode, space-charge-induced temporal broadening of the electron pulse with increased UV intensity was observed. Measurements of the electron pulse width were accomplished by operating the diffraction system as a streak camera with the streak voltage supplied by a laser-activated Cr:GaAs photoconductive switch.^{16,17} For conditions employed in Fig. 32.19, the electron pulse width used to obtain a RHEED pattern was measured at ~ 200 ps. The number of electrons delivered to the specimen per pulse was $\sim 5 \times 10^5$. As seen from Fig. 32.18(a), good RHEED patterns could be obtained with even a single electron pulse, making the study of nonrecurrent events possible. Better-quality diffraction patterns could be established by averaging several pulses, as shown in Fig. 32.18(b). Extension of the present technique to a few picosecond, or even subpicosecond, resolution should be possible by pulse compression of the driving laser and by minimization of electron temporal broadening.^{18,19}

In summary, we have demonstrated the technique of picosecond time-resolved-reflection, high-energy electron diffraction. This technique represents a direct time-resolved surface structural probe limited to the first few monolayers and synchronized with the driving laser source. Application of this technique could include the study of surface structural and chemical changes. We are currently constructing a picosecond RHEED system operating in ultrahigh vacuum. Future experiments will include the study of surface phase transformations and chemical reactions.

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