A picosecond electron gun for surface analysis

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I. INTRODUCTION

The ability to observe atom/surface dynamics is possible with the advent of electron-scattering techniques having picosecond and subpicosecond temporal resolution. Detailed studies of surface phase transformations and the dynamics of molecular interactions at surfaces are important in characterizing the initial stages of epitaxy during film growth, as well as the initial stages of surface melting/sublimation during the heating process. The reduced symmetry present at the surface often changes the energetics and time scales of important fundamental physical processes such as melting which emphasize the importance of understanding the role that the surface plays in initiating physical phenomena.

The combination of high temporal resolution light activated probes with surface sensitive analysis techniques provides a viable method for studying dynamics at surfaces with picosecond and even femtosecond temporal resolution. For example, time-resolved photoemission spectroscopy\(^1-3\) utilizing the pump-probe method has already proven to be a very useful technique for studying the electronic properties of surfaces and interfaces into the femtosecond time scale. The vibrational relaxation of adsorbates has been investigated using time-resolved sum frequency generation\(^4\) and infrared reflection absorption spectroscopy.\(^5\) Time-resolved second-harmonic generation\(^6,7\) has been used to study the structural properties of laser heated surfaces.

Here, we describe our efforts at combining reflection high-energy electron diffraction (RHEED) with ultrashort pulsed lasers. Time-resolved RHEED can be used to effectively monitor the surface structure and mean vibrational amplitude of atoms at the surface with picosecond time resolution.\(^8,9\) Specifically, we utilize a photoactivated RHEED electron gun excited by two picosecond laser pulses. The resulting electron-beam pulses are less than 5 ps wide, contain enough electrons, and can be sufficiently well focused to produce high quality electron-diffraction patterns. We describe the electron gun, the laser system, and the time-resolved diffraction system performance.

II. ELECTRON GUN DESIGN

A. Design methodology

The development of femtosecond lasers and their extensive use to study various physical phenomena have motivated the development of a subpicosecond temporal resolution streak camera. In a streak camera, an optical signal is converted into an electrical signal using a photocathode. Ideally, the resulting electron pulse should be an exact temporal and spatial replica of the optical excitation pulse. The electron pulse is rapidly scanned (streaked) across a two-dimensional observation plane, usually a phosphor screen/image intensifier\(^10\) or microchannel plates,\(^11\) by a set of high-speed detection plates.

Streak cameras operating in the subpicosecond time range have been successfully developed.\(^10,12-18\) The instruments attaining the high temporal resolution have optimized electron optical components. The achievement of high temporal resolution is the result of using a photocathode with an extremely narrow (photo)emission electron energy distribution. Narrow energy widths insure that initial electron velocities are nearly identical such that the adverse effects of chromatic aberrations can be minimized in the electron-beam forming optics. Photoemission electron energy widths as narrow as 0.1 eV can be achieved by selecting a photocathode material with a work function that is slightly less than the laser excitation photon energy, and employing a thin film which minimizes the redistribution of electron energies due to scattering during the process of emission and escape from the surface itself. The photocathode must be a conductor, or operated in a regime which guarantees charge balance to...
prevent the adverse effects of charging. Once a narrow temporal distribution of electrons has been produced with nearly the same kinetic energy, charged particle lenses must transport them to the target without temporally broadening the electron distribution. The primary causes of temporal redistribution in electron beams are path-length differences between the photocathode and target for different segments of the electron beam and space-charge (Boersch) effects. In order to minimize the effects of space-charge broadening, the electrons should be accelerated as quickly as possible once exiting the photocathode, and the number of (low-energy) beam crossovers should be minimized. In addition, the deflection plates used in streak cameras possess fringing field inhomogeneities which must be carefully controlled in order to reduce temporal dispersion. Further care must be taken in the design of the deflection plates to control capacitive effects which limit slew rates and switching speeds. Finally, the efficiency of the electron detector must be as high as possible. Single electron detection can be accomplished with phosphor screens and image intensifiers, or array detectors such as are used in a Vidicon.19

In order to minimize transit-time broadening due to path-length differences between electrons following different trajectories from the photocathode to the target, the initial electron spatial distribution must be small (much smaller than the inner diameter of the smaller electron focusing electrode). The use of long focal length lenses also decreases transit-time dispersion due to path-length differences. Space-charge broadening of the electron pulse near the photocathode can be minimized by using as large an extraction field as possible. The large extraction field quickly accelerates the electron pulse, thereby minimizing the time when low-energy electrons in close proximity can exchange energy. The extraction field is limited, however, by the vacuum breakdown field and the presence of any asperities on the cathode surface which might begin to undergo field emission. Typically, maximum dc electric fields near the cathode are limited to 5 kV/mm even though vacuum breakdown occurs near 30 kV/mm. Space-charge broadening can also occur in the postcathode electron optical column near beam crossovers and in regions of space where the electrons are significantly decelerated for focusing. The effects of space-charge broadening in the electron optical transport column can be mitigated through careful electron optical design of electrostatic lens elements12,14,18 or through the use of magnetic focusing lenses15 where the electron velocities are not changed. Both approaches have yielded picosecond temporal resolution, although the suitability of magnetic focusing systems to ultrahigh vacuums maintained for surface science remains problematic. Single-shot temporal resolution of 0.3 ps has been demonstrated14,16,17 For multiple-shot operation, jitter represents a major source for loss of temporal resolution. Synchroscan operation of a Photchron IV streak tube has demonstrated 930 fs temporal resolution.18 Our goal was an electron gun similar to the Photchron IV streak tube, but with focusing conditions that provide a real image 500 mm from the photocathode, and a beam that is relatively collimated (convergence angle about 1 mrad). Both requirements are necessary to use the electron gun for diffraction techniques, i.e., in order to separate diffraction spots and lines in the diffraction pattern.

### B. Electrode configuration

Primarily for design simplicity, and ultimately for UHV compatibility, we have selected an all electrostatic focusing system for our time-resolved RHEED gun. A schematic cross section of the photoactivated RHEED gun is shown in Fig. 1. The photocathode is held at potential $V_0$. While the emitted electrons are spatially divergent, a pinhole is used to extract relatively collimated electrons. This extraction electrode is part of the first lens and is held at a potential $V_1$, such that the photoelectron extraction field is large. Electrodes with potentials $V_1$, $V_2$, and $V_3$ form the first extraction lens. This zoom lens configuration is selected for its robust performance at long focal lengths, and small intrinsic aberrations.20 Electrodes with potentials $V_3$, $V_4$, and $V_5$ form a long focal length Einzel lens used for secondary focusing. Two lenses are necessary for focusing the electron beam at a distance of 0.5 m from the photocathode with a nominal spot diameter on the target of less than 300 μm with a nearly parallel beam. The requirement of a well focused and collimated electron beam is necessary for RHEED applications. In addition, no crossover points are located inside the electron lens column. These conditions are consistent with maintaining low electron density inside the electron lens column in order to minimize space charge broadening in that region. Some compromise between these requirements and that of maximizing the electric field and potential throughout the photocathode-to-anode region is incorporated in the design. For example, a relatively long electron lens column was necessary in order to achieve good electron collimation over the relatively long distance of 0.5 m.

The photocathode is composed of a 40 nm thick Ag film deposited ex situ on a sapphire substrate. When excited by a frequency doubled dye laser with 4.0 eV photon energy, the photoelectrons have a narrow distribution of electron energies. Although the photon energy is less than the work function of Ag (4.3 eV), the photoemission current displayed a linear dependence on the incident radiation intensity, indicat-

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**FIG. 1.** Electrode configuration of the photoactivated electron gun together with the nominal operating voltages.
ing that the emission process results from single photon processes. The lower- ing of work functions of thin metal films,\textsuperscript{21} thermal distribution of electron energies near the Fermi energy, or the presence of a thin layer of contamination on the surface may all play a role in initiating photoemission in this system with 4.0 eV photons. Since 40 nm of Ag is continuous and well conducting, surface charging of the photocathode cannot contribute to the reduction of the energy needed for photoemission. The spatial distribution of emitted electrons is defined primarily by the diameter of the illuminating radiation, and secondarily by the angle defining pinhole aperture in the extraction electrode $V_1$. The nominal diameter of this aperture is 200 $\mu$m. The size of the optical pinhole image formed on the photocathode determines the size of the focused beam on the screen and therefore represents the physical source of the electron optical transport lenses. Since the cathode is held at $-15$ kV, and the extraction electrode is held at $-8.75$ kV, a planar extraction field of 3.125 kV/mm accelerates the photoemitted electrons. Characteristic of planar extraction fields is a linear magnification of nearly 1. The angular width of the source at the pinhole is 10 mrad assuming uniform emission at the photocathode in all directions. This is a conservative upper limit.

The inner diameter of all of the lens elements is 15 mm with the exception of the inner diameter in the focusing electrode $V_2$ which is 3 mm. The zoom lens forms a virtual image of the source which is in turn focused onto the phosphor screen by the Einzel lens.

**C. Calculated electron trajectories and field distribution**

The electron optical properties of the RHEED gun were computed numerically. The potential and field distribution were found by numerically solving Laplace’s equation for given boundary conditions, using the boundary element method.\textsuperscript{22-25} The suitability of these techniques for the computation of paraxial properties and primary aberration coefficients for electron optical systems (in the absence of space charge) is widely accepted\textsuperscript{23,25} Specifically, cylindrically symmetric electrode geometries were approximated by straight-line segments, or electrode subelements. The two-dimensional Green’s function was integrated over the discretized electrode distribution, using pulse weighting functions for the charges, and either pulse-, or $\delta$-function,\textsuperscript{26} testing functions. The all orders electron trajectories were computed by integrating the equations of motion in time.\textsuperscript{27} The integration routine was based on a modified Adams–Moulton–Bashforth (predictor–corrector) method.\textsuperscript{28} In our implementation, both paraxial and all orders trajectories (where no approximations to the lens fields were made) were computed. Accurate calculations of third- and fifth-order geometric and first-order chromatic aberration coefficients were possible. The electron optical properties of each of the two lenses are summarized in Table I. The object $f_0$ and image side $f_1$ side focal lengths are given in millimeters. The object $F_0$ and image $F_1$ side focal points are referenced to the cathode ($z=0$ mm) and are also given in millimeters. The asymptotic projector aberration expansions for the spherical

<p>| Table I. Electron optical lens parameters. |</p>
<table>
<thead>
<tr>
<th>$V_0$ (kV)</th>
<th>$V_1$ (kV)</th>
<th>$V_2$ (kV)</th>
<th>$V_3$ (kV)</th>
<th>$V_4$ (kV)</th>
<th>$V_5$ (kV)</th>
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<td>-15.00</td>
<td>-8.75</td>
<td>-1.50</td>
<td>-2.50</td>
<td>-9.375</td>
<td>0.00</td>
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<p>| First-order optical properties |</p>
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<tr>
<th>$f_0$ (mm)</th>
<th>$f_1$ (mm)</th>
<th>$F_0$ (mm)</th>
<th>$F_1$ (mm)</th>
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</thead>
<tbody>
<tr>
<td>Zoom lens</td>
<td>39.92</td>
<td>56.48</td>
<td>-44.41</td>
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<tr>
<td>Einzel lens</td>
<td>111.00</td>
<td>124.21</td>
<td>-114.66</td>
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</table>

<p>| Spherical aberration coefficients |</p>
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<thead>
<tr>
<th>$C_{s0}$ (mm)</th>
<th>$C_{s1}$ (mm)</th>
<th>$C_{s2}$ (mm)</th>
<th>$C_{s3}$ (mm)</th>
<th>$C_{s4}$ (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zoom lens</td>
<td>6850.0</td>
<td>-23100.0</td>
<td>23900.0</td>
<td>-16600.0</td>
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<tr>
<td>Einzel lens</td>
<td>23800.0</td>
<td>-89000.0</td>
<td>128000.0</td>
<td>-81400.0</td>
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</tbody>
</table>

<p>| Chromatic aberration coefficients |</p>
<table>
<thead>
<tr>
<th>$C_{c0}$ (mm)</th>
<th>$C_{c1}$ (mm)</th>
<th>$C_{c2}$ (mm)</th>
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<tr>
<td>Zoom lens</td>
<td>62.8</td>
<td>-106.0</td>
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<tr>
<td>Einzel lens</td>
<td>273.0</td>
<td>517.0</td>
</tr>
</tbody>
</table>

$C_s$, and chromatic $C_c$, aberration coefficients\textsuperscript{27} are given in millimeters, where

$$C_s(\text{object}) = C_{s0} + \frac{C_{s1}}{M} + \frac{C_{s2}}{M^2} + \frac{C_{s3}}{M^3} + \frac{C_{s4}}{M^4},$$

and $M$ is the linear magnification of the lens. The performance of the entire electron optical transport system is given explicitly in Table II where each individual contribution to the final spot size is analyzed. Clearly, the spot size is dominated by the magnification of the source, implying that the RHEED spot on the target could be further reduced in size by using a smaller pinhole aperture (at the expense of current). Electron lenses possess focusing defects much in the same way that conventional optical lenses do. However, these focusing defects or aberrations are far more severe in

| Table II. Electron optical column performance. |
| --- | --- |
| Linear magnification | 1.68 |
| Angular magnification | 0.38 |
| Spherical aberration coefficient | 51859 mm |
| Chromatic aberration coefficient | 21 mm |
| Astigmatism and field curvature | 2671 mm |
| Coma | 46 mm |
| Distortion | 0.25 mm |
| Image radius (spherical aberration) | 0.0229 mm |
| Image radius (chromatic aberration) | 0.0011 mm |
| Image radius (magnified source) | 0.1680 mm |
| Image radius (astigmatism) | 0.0020 mm |
| Image radius (coma) | 0.0492 mm |
| Image radius (distortion) | 0.0066 mm |
| Image radius (diffraction) | 0.0000 mm |
| Image diameter (0.2 mm aperture) | 0.34 mm |
| Convergence angle (upper limit) | 3.80 mrad |
| Convergence angle (half intensity) | 1.30 mrad |
charged particle optics lenses. Of the primary aberrations, perhaps the most important is spherical aberration. The spherical aberration results from off-axis fields being stronger than those on axis, resulting in a stronger focusing of far off-axis trajectories. It is impossible to eliminate spherical aberrations in conventional, axially symmetric lenses without introducing space charge on axis. The chromatic aberration results from the focus defect associated with finite-energy distributions within the electron beam itself. Since electron lenses essentially focus the momentum of the electrons, more energetic electrons are focused more weakly than lower energy electrons, resulting in a focusing error. Coma is a focus defect whereby off-axis rays entering the optical system with differing angles are not uniformly focused. Characteristic coma-shaped beam distributions are common in systems dominated by coma. The distortion aberration causes off-axis to be displaced. The most common manifestation of distortion is the mapping of a square array into a barrel-shaped or pincushion-shaped pattern in the image plane. The image is perhaps the most important is spherical aberration. The matism and field curvature are aberrations whereby different axes of the image are focused with different strengths as a result of off-axis illumination. The resulting detoc in the image results. We summarize these third-order aberrations for our electrostatic lenses in Table II.

The temporal resolution of this lens column was also computed from a knowledge of the all orders trajectories. The average transit time from photocathode to the scattering target is 7350.7 ps. The deviation in transit times weighted for all trajectories through the RHEED gun was computed to be 1.26 ps, full width at half maximum. The choice of lens potentials shown in Table I indicates that the electron beam never has less than 5.625 keV of kinetic energy after the extraction pinhole in order to reduce the effects of space-charge broadening. This would have been impossible to achieve with a single electron lens column.

D. Electron gun construction

An electron gun has been constructed based on the above design. The outside diameter is 25 mm and the overall length is 330 mm. The lenses, anode, and diffraction plates are made out of aluminum and fabricated in our laboratory. Ceramic balls were used as spacers between the lenses. Deflection plates are incorporated at the end of the gun in order to steer the photoelectron beam towards the sample. The electron gun assembly is shielded with a μ-metal tube to avoid any distortion of the electron beam from stray magnetic fields (primarily the earth’s field and the magnets of the ion-pump system). The photocathode is a 40 nm thick Ag film evaporated on a sapphire window. Metals are good candidate materials for laser-driven photocathodes because of the ease of preparation, high damage threshold, and large free-electron density. The main drawbacks of metals are low quantum efficiencies and the high work function (4-5 eV) which require UV photons for linear (one photon) photoemission. The photocathodes are processed by thermal evaporation in a separate chamber and were installed into the electron gun without further processing.

Figure 2 shows the photocurrent as a function of the Ag film thickness measured with a Faraday cup. As a light source we used an Hg(Xe) arc lamp and a bandpass filter in order to separate the same wavelength as that of our Nd:YLF laser system ($\lambda = 4.2 \text{ eV}$). Since the photocathode is used in the transmission mode, the photoelectron current is expected to be governed by the photon and the electron mean-free path. The measured value of the photon mean-free path (penetration depth) is 211 Å for $h\nu = 4.2 \text{ eV}$ (Ref. 29) and the measured mean-free path for emission of these low-energy photoelectrons is about 10 Å. The result of three different series of film preparation shows that the photoyield is indeed decreasing with increasing silver film thickness (d > 20 nm). However, the actual behavior shows a very complex dependence of the photoelectron yield on film thickness with a second maximum at about 40 nm. Although the reason for this second maximum is not well understood, a change of the Ag film structure—such as the generation of colloidal silver features on the sapphire window—after a certain deposition thickness may account for this observation. Microscopic roughness of metal films—especially silver films—is known to enhance a variety of optical scattering processes at the surface and is well studied using different techniques such as surface enhanced Raman scattering. The major contribution to the enhancement is understood to originate from the coupling of photons into surface plasmons in regions of surface roughness. The decay of these nonradiative surface plasmons into one electron excitation can drastically increase the photoyield as first reported by Endrit and Spicer. The choice to use a 40 nm thick Ag film as a photocathode instead of a 20 nm thick one was made in order to assure good electrical conductivity across the film and to increase its durability to withstand many months of operation.

III. LASER SYSTEM

The laser system used in the experiments consists of a cw pumped, mode-locked Nd:YLF (neodimium yttrium lithium fluoride) master oscillator, a Nd:YLF regenerative amplifier, and a dual jet, synchronously pumped dye laser with two amplification stages (Fig. 3).

In particular, the output of the master oscillator (15 W, 1053 nm, 60 ps FWHM at 76 MHz) is split using a half-
FIG. 3. A schematic diagram of the laser system.

wave plate and a polarizer. A small fraction is used to seed the Nd:YLF regenerative amplifier and the rest is focused into a heated KTP crystal for second-harmonic generation. The frequency doubled output from the nonlinear crystal at 527 nm (1 W) is used to synchronously pump a dual jet dye laser with Rhodamine 6G as the gain medium and DODCl as the saturable absorber. The dye laser output (0.1 W, 76 MHz, and 800 fs FWHM) is then amplified in two Rhodamine 610 amplifications stages.

The dye amplification stages are pumped by the frequency doubled output of the regenerative amplifier. The amplifier is seeded with pulses from the main oscillator and a Faraday isolator is used to prevent feedback between the cavities. A lithium niobate Pockels cell (Medox E-O) is used to extract the amplified pulses from the regenerative amplifier cavity. The output (1 W, 1053 nm, 500 Hz) is frequency doubled in a BBO crystal to generate 0.5 W at 500 Hz (1 mJ) pulses at 527 nm. A diachronic beamsplitter is used to separate the fundamental from the second-harmonic output. The amplified pulses at 527 nm are then delayed and split with 8% of the power used in the first dye amplification stage and 92% in the second stage. A 27 cm focal length lens is used to focus the 527 nm pump pulses as well as the 590 nm pulses from the dye laser into the first amplification stage. A 27 cm focal length lens is used to focus the 527 nm pump pulses as well as the 590 nm pulses from the dye laser into the first amplification stage. The spot sizes at the first stage are 140 μm (dye laser) and 160 μm (pump), yielding an intensity of approximately 10^10 W/cm^2. A 10 cm focal length lens collimates the output of the first stage into the second stage where the dye laser spot size is 1 mm. The remaining 92% of the pump pulses is suitably delayed and focused into the second stage with a 1 m focal length lens. The amplification stages consist of 2 mm path-length cuvettes adapted to allow the flow of a solution of Rhodamine 610 in methanol. The concentration of Rhodamine 610 is adjusted to optimize the amplified power. The amplified dye laser output is 20 μJ/pulse at 500 Hz at 590 nm and 2 ps (FWHM).

The amplified dye laser pulses are then focused into a KDP crystal to generate the second harmonic at 295 nm, which are then focused onto the photocathode of the electron gun.

IV. ELECTRON GUN PERFORMANCE

The electron gun was tested first in a separate glass vacuum chamber with a base pressure of 10^-7 Torr. The operating voltages are shown in Fig. 1. A sapphire window passed the slightly focused (f=500 mm) UV beam into the vacuum system onto the Ag photocathode at normal incidence. Figure 4 shows the profile of the electron beam at the phosphor screen, 500 mm away from the cathode. This geometrical arrangement is identical to that used for RHEED. The pinhole following the cathode has a diameter of 200 μm. The measured full width at half maximum (FWHM) of the electron beam at the phosphor screen is 272 μm. If the photocathode aperture is the beam defining aperture, then the image profile should have been about 330 μm (see Table II: linear magnification −1.68). The fact that the measured profile was less than this indicates that the spot was limited by the source (incident light beam profile) and not the cutoff aperture. The convergence of the electron beam is about 1 mrad as measured by scanning the beam with the edge of a razor blade. This relatively small value ensures that the beam size is below 1 mm at the phosphor screen as well as on the sample. Therefore the quality of the diffraction pattern is dominated by the smoothness of the sample surface and not by the convergence (collimation) of the electron beam.

The average photocurrent was measured with a Faraday cup connected to a Keithley picocammeter. The log–log plot in Fig. 5 shows the linearity of the average electron current (slope=1) as a function of the laser-pulse energy. The current was found to be directly proportional to the laser intensity up to a pulse energy of about 0.5 μJ, focused to a 300 μm spot. The measured average electron current at this pulse energy was 3.5 pA, which results in about 5×10^4 electrons per pulse at a repetition rate of 500 Hz. Thus the nonaveraged current within individual pulses is 1.3 mA. Space-charge effects at higher intensities limit the emission of electrons as seen in Fig. 5 for I>0.5 μJ. When the charge of the extracted photoelectrons reaches a certain value q_e, the amplitude of the
electric field falls to zero and any additional photoelectrons cannot be emitted until part of the electron cloud has left the surface region. The time resolution is known to deteriorate quite rapidly above this region due to space-charge broadening. Therefore the electron pulse must be generated with minimum of space-charge dispersion in order to obtain the desired temporal resolution. The laser induced damage threshold of the photocathode is about 1 μJ (1 mJ/cm²) as evidenced by visible damage to the cathode.

In order to determine the temporal width of the electron pulse, we use a jitter-free streak camera device. An ultrashort voltage ramp pulse with a rise time of a few hundred ps on a pair of deflection plates is synchronized with the electron pulse and sweeps it across the phosphor screen. The duration of the electron beam is proportional to the calibrated length of the streak. This sweep requires the electrical ramp pulse (5 kV) to be synchronized with the electron pulse with high accuracy. In the repetition mode, the streak length, and therefore the overall time resolution of the electron pulse measurement, is limited by the jitter between the high-voltage pulse for the deflection plate ramp and the electron pulse. Conventional electronics can at best offer a jitter of several ps for short times, in addition to long term delay drift. Instead, we use a laser activated photoconductive switch as developed by Mourou et al. A diagram of the experimental arrangement is shown in Fig. 6. The laser light is split into two beams. One beam is frequency quadrupled and used to generate a pulse of electrons at the photocathode of the electron gun. The other beam is suitably delayed and then used to strike a GaAs photoconductive switch which triggers a fast RC circuit to begin charging the pair of deflection plates to a potential of 5 kV. The switching is based on the photogeneration of carriers in a semiconductor bulk by the ultrashort laser pulse. The rise time of the switch is essentially determined by pulse duration and circuit bandwidth. The sweep time of the voltage ramp is given by the RC circuit, which includes R and the capacitance of the deflection plates Cplates.

We could not experimentally verify the limit of the temporal resolution because of the nonlinearities in the sweep speed of the GaAs switch. According to our preliminary results, an upper limit on the electron pulse width of τ<5 ps can be concluded. Further work is underway to calibrate the GaAs switch and the optical setup to overcome this problem.

V. TIME-RESOLVED ELECTRON DIFFRACTION

A. RHEED setup

Reflection high-energy electron diffraction (RHEED) is one of several diffraction techniques that have the capability of investigating surface order with great accuracy. In the RHEED technique, high-energy electrons (10–20 kV) are diffracted off an ordered surface at a glancing angle (2°). For a flat single crystal, the RHEED pattern consists of a set of streaks oriented perpendicular to the sample surface (elastic two-dimensional Laue scattering) as shown schematically in Fig. 7.

The intensity of these diffraction patterns is temperature dependent, as in low-energy electron diffraction. As the temperature is raised, the atomic vibrational amplitude increases, which leads to a reduction of elastically scattered electrons and causes a reduction of the diffraction pattern intensity. This is known as the Debye–Waller effect. Figure 8(a) shows scans through one of the RHEED streaks from a platinum single crystal taken at various temperatures, obtained during a static heating experiment. The measured decay of the peak height as a function of temperature is shown in Fig. 7. From the linearity of this semilogarithmic plot, it is
evident that the RHEED streak intensity follows an exponential decay in accordance with the Debye–Waller theory.\textsuperscript{35} These intensities give us an accurate calibration of the temperature of the surface, i.e., the temperature of the surface can be determined by comparing the pattern intensity with this calibration curve.

The use of time-resolved RHEED to study ultrafast dynamics at solid surfaces is a fairly recent development. It is explained in detail by Elsayed-Ali \textit{et al.}\textsuperscript{8} and will be discussed here only briefly. The basic concept of this technique is the use of a photoactivated electron gun, which produces the electron probe pulse synchronized to a pump laser pulse to monitor a transient event. A schematic diagram of the experimental setup is illustrated in Fig. 9. The amplified dye laser light is split into two beams. One beam (pump) is used to transiently heat the sample surface. The other beam (probe) is frequency doubled and suitably delayed, and is used to generate a pulse of electrons at the photocathode of the electron gun. The electrons are then used to probe the structure and mean vibrational amplitude of the surface by measuring the intensity change of the streaks on a phosphor screen as a function of the pump-probe delay. This enables the observation of laser induced transient temperature and structural changes at the surface region with a time resolution limited by the duration of the electron pulse.

### B. Electron detection system

In order to obtain quantitative information of the diffraction intensity, time-resolved RHEED is signal limited when compared to conventional static RHEED systems. The electron signal has a pulse length of only a few µs and the number of electrons available in a single pulse is small ($\sim 10^4$ per pulse). Therefore it is essential to have an image detecting device with sensitivity to single electrons with a very low dark current. This requirement can be fulfilled with high gain microchannel plates (MCP) as an amplification stage followed by a phosphor screen. The signal obtained from the phosphor screen can then be recorded and analyzed. The spatial resolution is limited by the MCP channel diameter and their center-to-center spacing.

As shown in the schematic diagram of Fig. 10, we use a Chevron MCP detector with a 40 mm diameter P20 phosphor screen on a fiber-optic faceplate as the vacuum interface. The fiber-optic faceplate provides one-to-one imaging of the diffraction pattern, i.e., a recording film or an array detector with another fiber-optic window can be placed directly in

![Fig. 9. Experimental setup for time-resolved RHEED. This technique allows one to observe laser induced transient temperature and structural changes at the surface region with a time resolution given by the duration of the electron pulse.](image)

![Fig. 10. Electron detection and data collection systems currently used in our time-resolved RHEED setup.](image)
Thus there are a number of choices that we made in our system configuration to achieve high performance as well as low cost. Our time resolution comes from the timing between laser and electron pulses (time when the laser beam impinged on the sample and the time when the electron beam arrived). Because the laser repetition rate is 500 Hz, no attempt is made to synchronize the laser to the video camera. Instead, multiple pulses (from 80 to 500) are averaged for each video frame. Further signal averaging is accomplished by adding data from multiple video frames.

For our CCD detector we use a Pulnix TM-745E camera. This was chosen because of its relatively low cost, high sensitivity, as well as its ability to integrate low light level images on the camera. Typically we would integrate each frame for 1 s. This is accomplished by using a logic signal from a Keithley P10-12 board in the computer to turn on the Image inhibit line to the camera. The only disadvantage of this camera is its inability to manually control the black level. In most cases this is not a serious problem since the background level is determined from video frames collected with the laser on and with the laser off. These are collected alternately by opening and closing a shutter in the path of the laser beam by using a logic signal from the Keithley board to a Uniblitz VSI1451W0 control unit and shutter.

We use an Imaging Technology VFG frame grabber. Though we had planned to use a feature of this frame grabber board that allowed the use of a higher performance non-standard video camera, we found that the Pulnix camera worked adequately for our application. The data collection cycle is designed to minimize deadtime. At the end of a camera integration cycle the whole video frame is grabbed. The shutter is opened or closed and the next camera integration cycle is started. During the time the camera is integrating the next frame, the data from the previous frame is processed. Thus it is crucial that all the data can be processed by the time the next video frame from the camera is ready to read. We found that for a 1 s integration time more than half of the video frame could be processed. A greater proportion of the frame could be handled either by increasing the integration time or by using a faster computer.

We organized the data collection into different regions on the CCD array. Usually one or more regions are used to collect the RHEED pattern, one to measure the overall background level, and a few to measure the intensity in individual streaks. The most time consuming portion of data processing is the regions consists of transferring the data values from the frame grabber board memory into the computer memory. Then the data can be stored as an image.

Usually, the data in a region is compressed into line formal by adding the data values by row and/or by column. This generates a line scan that is typically used to show the multiple peaks in a RHEED spectrum. Also, the total intensity in the region is computed and stored. Such data is analogous to RHEED oscillation data that is used in MBE growth. The total intensity is displayed on the computer screen primarily to allow the users to follow the data collection process, but it is also useful for measuring the background level and for calibration. The whole video image is also displayed on a separate TV monitor. In order to achieve adequate signal-to-noise ratio, multiple video frames are collected and averaged for each time delay. At the end of signal averaging for a given time delay (typically 2 min) the laser-on and laser-off data for each region is stored on the hard disk, the laser delay line is changed to a new time delay, the data arrays are reinitialized, and the process is restarted. By storing the laser-on and laser-off data separately, further data processing could be delayed until after the experiment is completed.

D. Performance

As mentioned above, the electron pulse is used for surface analysis, primarily for probing the surface temperature and order by utilizing the time-resolved RHEED technique. Figure 11 shows the two-dimensional diffraction pattern (RHEED streaks) of a Ni(100) surface with adsorbed oxygen c(2×2) structure, along [010] azimuth, using ultrashort (τ<5 ps) electron pulses. The image was recorded on a photographic film in contact with the fiber-optic faceplate. The electron gun was operated at 18 kV. The fine streak pattern (zero-order Laue ring L0) and even some Kikuchi lines are clearly visible and demonstrate the excellent signal-to-noise ratio that we achieve with our short electron pulses. The first-order Laue ring L1 does not appear in the picture because the L1 radius for the incident electron-beam energy of 18 keV is larger than that of the RHEED screen. As previously observed by several authors,37 adsorption of oxygen on clean Ni(100) at 400 K leads to the appearance of an ordered...
FIG. 12. Line scan through the diffraction patterns of a Pt(ll1) surface. (a) The raw data after signal averaging for 60 frames at 1 s, (b) after subtracting the CCD background from the raw data.

overstructure. First, at low oxygen coverage ($\Theta < 1/4$ ML) the overstructure forms the $p(2\times2)$ structure and then, for longer exposure ($\Theta \approx 1/2$ ML), the $c(2\times2)$ structure forms. It should be emphasized that in spite of the low oxygen coverage and the small number of electrons produced by the ultrashort electron pulses (compared to the conventional cw RHEED technique) the additional diffraction pattern due to the ordered oxygen overstructure is clearly visible. This allows investigation of transient phenomena of the substrate and adsorbed species at the same time. For example, a thermal nonequilibrium in the adsorbate–metal complex after a laser heating pulse can be directly monitored in real time. Initial results will be discussed in a later paper.

Figure 12 shows a line scan through the diffraction pattern of a Pt(ll1) surface approximately perpendicular to the streaks. The data were obtained with the CCD camera and processed with the frame grabber as discussed above. The line scan is an integration of five rows (five pixels of the CCD camera) in order to average over fluctuation in the direction along the streaks. Figure 12(a) shows the raw data after signal averaging for 1 min (60 frames at 1 s). Immediately after the measurement, the CCD background of 60 frames was also measured and subtracted from the raw data. Figure 12(b) illustrates the same line scan after the background subtraction. No smoothing is necessary for further data processing.

As discussed above, the basic idea of the time-resolved RHEED experiment is to observe laser induced transient temperature and structural changes of the surface region. We measured the temperature change of the Pt(ll1) single-crystal surface after a 100 ps IR laser heating pulse. The reduced streak intensity due to the laser pulse was compared with a calibration curve obtained by a static heating measurement. Based on this calibration the heating IR laser pulse raised the surface temperature by 20 K (see Fig. 13). In order to minimize experimental error due to long term drift, a computer controlled shutter is placed in the path of the IR heating beam. The purpose of the shutter was to chop the IR beam with a frequency of 1 Hz which allows one to quickly alternate between a hot (shutter open) and a cold (shutter closed) measurement. The normalized difference between the hot and the cold measurement was averaged over 60 cycles. An exposure time for one point (delay time) of 2 min (includes on and off time) is needed for such a small temperature change to average out shot-to-shot laser fluctuations of the IR heating laser pulse, the electron probe pulse, and the gain of the MCP plates. Because of the glancing angle of incidence of the electron probe pulse and the perpendicular direction of the heating laser pulse, the temporal resolution is not only limited by the pulse width of the electron gun but also by the time difference between an electron scattered at the front edge of the sample to an electron scattered at the trailing edge of the sample. Therefore, in order to increase the overall temporal resolution of this technique below 15 ps, the sample has to have a width smaller than 1 mm.

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