A pulsed electron gun for ultrafast electron diffraction at surfaces

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The construction of a pulsed electron gun for ultrafast reflection high-energy electron diffraction experiments at surfaces is reported. Special emphasis is placed on the characterization of the electron source: a photocathode, consisting of a 10 nm thin Au film deposited onto a sapphire substrate. Electron pulses are generated by the illumination of the film with ultraviolet laser pulses of femtosecond duration. The photoelectrons are emitted homogeneously across the photocathode with an energy distribution of 0.1 eV width. After leaving the Au film, the electrons are accelerated to kinetic energies of up to 15 keV. Focusing is accomplished by an electrostatic lens. The temporal resolution of the experiment is determined by the probing time of the electrons traveling across the surface which is about 30 ps. However, the duration of the electron pulses can be reduced to less than 6 ps. © 2007 American Institute of Physics. [DOI: 10.1063/1.2431088]

I. INTRODUCTION

The dynamics of surface atoms subsequent to an electronic or vibrational excitation are of great interest for basic research as well as for applications in materials processing. Such an analysis on the atomic level requires both high spatial and temporal resolution. Because the relevant timescales for electron-phonon and phonon-phonon coupling are in the femtosecond (fs) to picosecond (ps) regime,1 the commonly used approach is a pump-probe setup using fs-laser pulses for excitation. The structural analysis of surfaces with sub-angstrom resolution can be accomplished by electron or x-ray diffraction. In contrast to other time-resolved surface sensitive methods such as sum-frequency generation or reflection high-energy electron diffraction (RHEED) or a reflection high-energy electron diffraction (LEED) or a reflection high-energy electron diffraction (RHEED) setup.2

As a consequence, ultrafast time-resolved electron diffraction (UED) in a pump-probe setup with fs-laser pulse excitation is a promising tool to study the dynamics of surfaces on an atomic level with a temporal resolution of ps to fs.3–10

In this article, we describe the construction of an electron gun used in RHEED experiments at crystal surfaces with a temporal resolution below 30 ps. In contrast to the most recent developments in the field,11 our gun is equipped with an electrostatic instead of a magnetic lens. This approach was chosen to ensure the ultrahigh vacuum required for long exposure times, which are necessary because of the low-electron reflectivity of surfaces (10−4). The achievement of an excellent vacuum, however, is often hampered by outgassing of isolation materials and the necessary water cooling of the magnetic lens. Compared to other pulsed electron guns with electrostatic lenses,5,12 our gun is more compact and avoids a crossover of the electron trajectories. These are important prerequisites for the generation of ultrashort electron pulses.

II. EXPERIMENTAL SETUP
A. General setup

A sketch of the time-resolved diffraction experiment is shown in Fig. 1. A 45 fs-laser pulse from a Ti:sapphire laser system (wavelength 800 nm) is split into two beams. The first traverses an optical delay line and is then used to excite the sample surface (pump). The second is frequency-tripled in a two-step process using nonlinear optical crystals ( β-barium borate, BBO ), resulting in ultraviolet (UV) laser pulses with a wavelength of 266 nm, (spectral width Δλ =2.8 nm, full width at half maximum (FWHM), 30 nJ/pulse). These UV pulses generate photoelectron bunches in the photocathode of the electron gun, which are accelerated up to 15 keV. The electrons impinge onto the sample surface under a grazing angle of incidence (αm=5°). The resulting electron diffraction pattern is amplified by a microchannel plate, visualized by a phosphor screen and recorded by a cooled charge-coupled device (CCD) camera. The time delay between the laser pump and the electron probe pulse can be changed by a variation of the delay line length. The setup was used with two different laser systems, one providing up to 2 ml/pulse at a repetition rate of 1 kHz, the other with 0.5 ml/pulse at a repetition rate of 5 kHz. Through integration over some 104 pulses, diffraction patterns with good statistics are recorded in both cases.

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B. The ultrafast electron gun

The setup of the ultrafast electron gun resembles a conventional fine-focus LEED gun \cite{13} with a photocathode instead of a filament and a Wehnelt cylinder. The gun elements consist of graphitized cylindrical titanium electrodes that are isolated by ceramic rings. The entire stack of electron-optical elements is mounted between two base plates as depicted in Fig. 2 (to scale). The setup has an overall length of 120 mm, i.e., only half as much as the gun described in Ref. 6. The electron pulses are generated via photoemission from a thin Au film (the photocathode), which is back-illuminated by fs-UV-laser pulses with a wavelength of 266 nm (4.65 eV/ photon). The UV light enters the electron gun through a magnesium fluoride viewport with a transmissivity for UV light, its high thermal conductivity and vacuum compatibility.

Prior to the deposition of the Au films, mechanically stable electrical contact pads, as shown in the inset of Fig. 3, were deposited onto the boundary of the sapphire substrate by thermal evaporation of a micrometer-thick Au film in high vacuum ($10^{-8}$ mbar) through a shadow mask.

The sapphire substrate was then mounted in a small vacuum chamber equipped with a magnesium fluoride viewport, a Faraday cup, and an e-beam Au evaporator, which had previously been calibrated by a quartz-microbalance. By biasing the Faraday cup and the photocathode with $+30$ V and $-30$ V, respectively, it was guaranteed that all photoemitted electrons would be detected by the Faraday cup. Au films with thicknesses up to 50 nm were deposited gradually onto the prestructured substrate at room temperature. After each deposition, the photocathode was back-illuminated by UV light from a Hg discharge lamp, and the resulting photocurrent was measured with the Faraday cup and a picomammeter. The results are plotted as a function of the Au film thickness in Fig. 3 for different photocathodes. The photocurrents have been normalized to the respective peak currents of typically 3 nA.

III. LASER-ACTIVATED PHOTOCATHODE

A. Au film thickness

As will be discussed in Sec. IV B, the electron pulse duration is essentially determined by the electrostatic repulsion of the electrons (space charge effects) and their initial energy distribution (vacuum dispersion). To reduce the latter, the initial energy distribution of the photoelectrons should be as narrow as possible. This can be accomplished if only electrons close to the Fermi level are photoemitted. Furthermore, the photocathode material must be inert because the photocathodes are exposed to ambient air during the assembly of the electron gun. The latter requirement is met by the choice of Au as the photocathode material, since it is highly inert. Additionally, the work function of 45 nm thin Au film was found to be 3.8 eV (Ref. 14) instead of 5.2 eV for bulk material. Therefore, by using UV light with a photon energy of 4.65 eV only electrons near the Fermi level are photoemitted. The ideal substrate is sapphire with its high transmissivity for UV light, its high thermal conductivity (compared to quartz or magnesium fluoride) and vacuum compatibility.
are emitted only from a surface layer with a thickness given by the electron inelastic mean free path by Seah and Dench. The decrease of the number of excited electrons due to inelastic scattering along their path to the surface, a simple expression derived from a simple model taking into account only light absorption and inelastic scattering of the excited electrons.

The photoemission yield under back-illumination is determined by two counteracting effects: For films thinner than the absorption length \( \alpha^{-1} \) of the UV light the photon absorption in the Au film increases with film thickness, leading to an increase of the photoelectron yield. For films thicker than the electron inelastic mean free path \( \lambda_e \), however, electrons are emitted only from a surface layer with a thickness given by \( \lambda_e \): With increasing thickness the emission reaches a maximum value and finally decreases with the absorption length \( \alpha^{-1} \) for UV light. Assuming the Lambert-Beer law for the attenuation of the UV light intensity and an exponential decrease of the number of excited electrons due to inelastic scattering along their path to the surface, a simple expression for the thickness-dependence of the photocurrent could be derived:

\[
I_e(D) = I_{0e} \frac{\alpha \lambda_e}{1 - \alpha \lambda_e} \left( \exp(-\alpha D) - \exp\left(\frac{-D}{\lambda_e}\right) \right),
\]

with the film thickness \( D \), a constant \( I_{0e} \), the absorption coefficient \( \alpha \) for the UV light, and the electron inelastic mean free path \( \lambda_e \). The solid line in Fig. 3 shows the result of Eq. (1) with the parameters \( \alpha^{-1} = 12 \text{ nm} \) and \( \lambda_e = 4.1 \text{ nm} \). These values are in agreement with published values of the optical constants of thin Au films and with the prediction of the electron inelastic mean free path by Seah and Dench. Although the model does not include the previously mentioned change of the Au work function, it provides a satisfactory description of the measured data. The highest photocurrent is found for a Au film thickness of approximately 8 nm from both the model and the experiment.

Because the photocurrent decreased in the course of longer exposures to UV light, an additional 2 nm thin W interlayer was added as an adhesive between the sapphire and the Au film to prevent dewetting. The photocathodes prepared in this way exhibit excellent long-term stability: The UED gun has been used with the same photocathode for more than two years without noticeable deterioration.

The energy distribution of the photoelectrons emitted form a Au photocathode with a few nanometers thickness was analyzed in a time-of-flight spectrometer (TOF). An infrared (IR) filter (Schott UG11) was used during illumination with the frequency-tripled laser pulses to eliminate the possibility of multiphoton photoemission. The pressure in the UHV chamber was in the low \( 10^{-9} \text{ mbar} \) range during the experiments. Because the sample holder did not allow back illumination, the measurements were conducted in a reflection geometry. Figure 4 shows the energy distribution of photoelectrons emitted from a photocathode which had been prestructured in the way explained in Sec. III A, while the thin Au film was deposited in situ (dashed line). The small peak at 0.16 eV binding energy originates from the W adhesive layer as confirmed by comparison with a spectrum recorded before the Au deposition (not shown here). Since the film thickness could not be controlled exactly, the Au film might be somewhat thinner than 10 nm. However, the narrow photoelectron energy distribution is also observed in the TOF spectrum of a 9 nm thick Au film on Si(111) (solid line in Fig. 4). Since published values for the work function of bulk Au are in the range of 4.9–5.4 eV (Refs. 15–17) a photon energy of 4.65 eV would be insufficient to generate photoelectrons in a one-photon process from bulk material. It is evident from the narrow energy distribution of less than 0.1 eV width that the work function of the thin Au films is lowered to about 4.5 eV and that only electrons close to the Fermi edge are excited above the vacuum level. A lowered work function of 3.8 eV has previously been observed for thin Au films. However, the reason for the less pronounced decrease of the work function in our study is still unclear and requires a more detailed investigation.

The result is transferable to the transmission geometry: Inelastic scattering events will either lead to a minor redistribution within the energy distribution or—in case of higher-energy losses along the paths of the excited electrons—to a reduced number of photoelectrons.
due to the higher average intensity of the Hg discharge images have been rescaled to a comparable grayscale representation.

After the deposition of the W layer, the majority of the photoelectrons is mainly determined by the beam profile of the UV light from a Hg discharge lamp prior to Au deposition [Fig. 5(a)], and with UV laser pulses after the \textit{in situ} deposition of a 7 nm thick Au film [Fig. 5(b)], respectively. Both images show the same area on the surface with a field of view of 100 μm. The images have been rescaled to a comparable grayscale representation. In fact, the image generated with the UV laser pulses is much darker indicating a smaller electron yield. The images show a homogeneous intensity distribution without visible charging of the substrate. Inhomogeneities, initially present on the surface, are partially overgrown by the Au film. We conclude that both the W adhesive layer and the Au film are continuous across the field of view within the spatial resolution of the PEEM instrument. The bright spots are interpreted as contaminations, adsorbed on the sample during the transfer from the preparation chamber into the PEEM after the deposition of the W layer. The majority of the photoelectrons (>99%) originates from the homogeneous areas. Due to the higher average intensity of the Hg discharge lamp, the signal-to-noise ratio is much higher in Fig. 5(a) than in Fig. 5(b). For the application in the UED gun, these results suggest that the spatial distribution of the photoelectrons is mainly determined by the beam profile of the UV laser pulses and that local concentrations of charge carriers can be ruled out.

C. Electron optics

Due to the very low energy spread of the photoelectrons ($\Delta E_{\text{kin}} = 0.1$ eV, see Fig. 4) the acceleration to high energies results in an almost parallel beam with a divergence of less than 0.4° for 10 keV electrons. Further beam divergence is caused by a bending of the equipotential lines at the anode aperture (see Fig. 6), which can be described in terms of a \textit{punch-through effect} of the electrostatic potential resulting in an electrostatic divergent lens. The electron trajectories are thus bent away from the optical axis, leading to a defocused beam. The electrostatic potential was calculated by a finite elements representation of the Laplace equation, which was solved with the successive over-relaxation technique.\textsuperscript{22} In addition, electron trajectories were simulated with SIMION.\textsuperscript{23} All simulations have been performed with an acceleration voltage of 10 kV. The SIMION simulations show that only electrons originating from an area with a diameter of 300 μm around the center of the photocathode pass through all apertures in the electron gun. With this restriction to the starting points of the electron trajectories, a divergence angle of 2.3° due to the deflection in the anode region was found. Therefore, the electron beam has to be focused. The application of a mesh instead of a single aperture was found to lower the angular divergence by almost a factor of 10 because of the reduced punch-through effect for the smaller apertures of the mesh compared to the single aperture. Such a mesh, however, would reduce the transmissivity by about 50%.

In this setup a crossover of the electron trajectories has to be avoided because it would lead to a high space charge and a severe temporal broadening. Therefore, a Wehnelt cylinder, which is usually found in conventional thermionic emission guns, has been omitted.

In the simulation, the focal point was found to be 60 cm away from the photocathode for a lens potential of $U_L = 0.6 U_{\text{PC}}$ with $U_{\text{PC}}$ being the photocathode potential. This result is in excellent agreement with the experimentally determined value of 0.58 $U_{\text{PC}}$ for an optimal focus at a distance of 55 cm between the photocathode and the microchannel plate.

IV. PERFORMANCE

A. Spatial resolution

To quantify the spatial resolution of the electron gun, the transfer width\textsuperscript{24,25} was extracted from the diffraction patterns of a Si(001)-(2 x 1) surface shown in Fig. 7. The images were generated with 8.58 keV electrons, the angle of incidence was 6°. As a source of UV light both a Hg discharge lamp [Fig. 7(a)] and 266 nm fs-laser pulses [Fig. 7(b)] were used. The zero-order Laue ring with integer order and (2 x 1) reconstruction spots is seen on the left side of the diffraction pattern. The first half-order Laue ring consisting of faint (2 x 1) spots is indicated in the right part of the pattern. Figure 8 shows line profiles along and across the (00)-spot
obtained from Figs. 7(a) and 7(b). The diffraction spots are sharper during the illumination with fs-laser pulses, while at the same time the signal-to-background ratio increases from 12 to 18. The improved resolution is caused by the very narrow energy spread of the photoelectrons during laser illumination (see Fig. 4) which results in an almost parallel electron beam after acceleration. Due to the higher photon energies delivered by the Hg discharge lamp up to 5.3 eV more electron states contribute to the photoemission yield, which leads to an initially broader angular distribution of the photoelectrons. This beam divergence cannot be compensated by focusing because the photocathode emits electrons from a large area and not from a point source. The resulting transfer widths during laser illumination are 53 Å perpendicular and 376 Å parallel to the plane of incidence. The beam diameter was determined by moving the sample across the beam and measuring the displacement between the sample positions at which the diffraction pattern vanishes. This procedure delivers a beam diameter at the sample of 380 μm. The electron beam’s angular divergence of 0.1° (1.6 mrad) follows from the diameter of the specular spot on the microchannel plate and the distance between sample and detector.

B. Temporal resolution

The temporal performance of the gun is demonstrated in a pump-probe experiment. A 10 nm thick epitaxial Bi(111) film on Si(001) has been used as a test system. If the sample is heated by ultrashort laser pulses (800 nm, 45 fs, 1.5 mJ/cm², 5 kHz), the transient intensity of the (00)-spot drops due to the Debye-Waller effect. 26, 27 The low absorption length of α⁻¹ = 13 nm for light at a wavelength of 800 nm together with a very low surface Debye temperature of θ⁰,Bi = 52 K ensures both a large temperature rise and a pronounced Debye-Waller effect, i.e., high sensitivity upon laser excitation. As shown in Fig. 9, the intensity decreases linearly to about 80% of the initial intensity which reflects a temperature rise from 90 to 150 K.

The temporal resolution in our surface sensitive RHEED experiment is dominated by the velocity mismatch between the excitation laser pulse at normal incidence onto the sample surface and the 7 keV electrons, traveling with 1/6 of the speed of light, at grazing incidence. 6 This leads to a time lag between electrons scattered from the leading and trailing edge of the sample. For 7 keV-electrons impinging onto a sample of 3 mm width under an angle of incidence of 5°, the resulting probing time is 60 ps. As a conservative measure for the temporal resolution in the UED experiment, one can determine the time needed for the intensity drop from 90% to 80%.
the electron beam significantly improves the temporal resolution of the experiment, can be reduced from 60 to 28 ps if only the sample edge is probed by the electron beam.

10% of the total intensity change. A reduction of the effective sample width by a partial retraction of the sample from the photocathode-anode region:

differences, as discussed in the following:

However, the electron pulse duration, which ultimately limits the temporal resolution, can be reduced to less than 6 ps by lowering the beam current per pulse. It is determined by space charge effects, vacuum dispersion and path length differences, as discussed in the following:

1. **Photocathode-anode region:** After emission from the photocathode, the electrons are accelerated from almost zero velocity to their final energy of \(eU_{PC}\). Due to the high extraction field, the electrons’ trajectories are nearly parallel in this region without a temporal spread due to path length differences. Following a model for short drift times by Qian and Elsayed-Ali,\(^{28}\) the space charge broadening accounts for less than 5 fs for 1500 electrons per pulse in a bunch of 300 \(\mu m\) diameter and an acceleration to 7 keV on a distance of 2 mm. Space charge effects are negligible in this region because the electrons travel only 80 ps from the photocathode to the anode. The main contribution is caused by the width of the initial energy distribution after photoemission. A width \(\Delta E\) results in a time spread \(\Delta t_{\text{acc}}\) according to

\[
\Delta t_{\text{acc}} = \sqrt{\frac{2m_e}{e^2} \frac{\Delta E}{|E|}}. \tag{2}
\]

With \(\Delta E=0.1\) eV (see Sec. III B) and \(|E|=3.5\) kV/mm, Eq. (2) yields \(\Delta t_{\text{acc}}=300\) fs. Instabilities of the acceleration voltage are negligible for the electron pulse broadening. Since the relevant contributions to these instabilities usually have time constants of some milliseconds, the electrons in a pulse are subject to an almost constant potential during the acceleration period of 80 ps.

2. **Drift region:** In contrast to the acceleration region, the instability of the beam energy has to be taken into account in the drift region between the anode and the sample surface since it leads to small differences in the travel times of successive electron bunches. The power supply used in these experiments is stabilized to less than 1 V,\(^{30}\) and thus the effect results in a contribution of 260 fs for a drift length of 185 mm and a resulting travel time of 3.7 ns. However, the time spread in the drift region is dominated by space charge repulsion due to the long travel time. The application of a simple model proposed by Siwick et al.\(^{29}\) gives a pulse broadening of 4.3 ps and an energy spread of 27 eV at the sample position, assuming 1500 electrons and an initial energy distribution of 0.1 eV width.

3. **Electrostatic lens:** Focusing in the electrostatic lens is accomplished by the fact that electrons traveling on trajectories farther away from the optical axis traverse a region of higher electric potential and are therefore decelerated and deflected back onto the optical axis. This adds two further contributions to the temporal spread: the electrons on the outer paths travel longer distances, and, in addition, are decelerated more strongly than those traveling along the optical axis. In the SIMION simulations, the sum of these two effects was found to be 900 fs for 10 keV electrons. Since the electron trajectories remain unaltered if the acceleration and the lens potential are changed by the same factor, the resulting contribution is 1080 fs for 7 keV electrons due to the lower electron velocity.

The electron pulse duration is estimated from the sum of the above-named contributions

\[
\Delta t_{\text{pulse}} = \Delta t_{\text{acc}} + \Delta t_{\text{drift}} + \Delta t_{\text{lens}} \approx 6 \text{ ps} \tag{3}
\]

for 1500 electrons per pulse at an acceleration voltage of 7 kV.

V. CONCLUSIONS

The construction of a pulsed high-energy electron gun for time-resolved RHEED experiments in a pump-probe setup on the basis of a conventional fine focus electron gun has been described. The filament was replaced by a photocathode consisting of a 10 nm Au film deposited onto a prestructured sapphire substrate. The illumination of the photocathode with fs-UV-laser pulses generates photoelectrons with homogeneous spatial distribution and a narrow initial energy distribution of only 0.1 eV width. The gun can be operated at electron energies between 0 and 15 keV. The divergent electron beam is focused by an electrostatic Einzel lens. RHEED diffraction patterns with good statistics were recorded in 80 s from a phosphor screen by a cooled CCD camera after an image intensification with a microchannel plate. The temporal resolution in this surface sensitive setup is dominated by the velocity mismatch between the electrons and the laser pulse. It depends linearly on the width of the sample and could be reduced from 60 to 28 ps by probing only the sample edge with the electron beam. However, the ultimate limit for the temporal resolution is given by the electron pulse duration, which can be reduced to about 6 ps by lowering the beam current.
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21 For more information refer to www.simion.com.