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LOW ENERGY ELECTRON DIFFRACTION WITH MICROSCOPIC RESOLUTION

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Abstract

We report on the development of a Scanning Low Energy Diffraction Microscope, operating in the range of 250 to 1000 eV primary energy. By discriminating against inelastically scattered electrons, low energy electron diffraction (LEED) patterns are obtained from areas of about 100 nm in size. By selecting a particular diffracted beam dark-field images of the surface structure are obtained in the scanning mode. Examples are given for polycrystalline Si and clean and adsorbate covered Si(111) surfaces.

KEY WORDS: Low energy electron diffraction, Ultra-High Vacuum Microscope, Scanning electron microscope, surface structure

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Introduction

Low energy electron diffraction, in the following called LEED, is a well established technique in surface science with numerous applications. They range from a simple check of the order of a crystalline surface to elaborate surface structure determinations in the presence of adsorbates. The position of adsorbate atoms relative to the substrate atoms can be determined as well as a possible reconstruction of the substrate caused by chemisorbed atoms or molecules. Clean surfaces have been shown to exhibit contraction and/or expansion of the layer spacings near the surface as well as lateral displacements of rows of atoms. The analysis of the profile of diffracted beams yields information on the nature and amount of defects at the surface. Examples of these and other applications may be found in the recent book by Marcus and Jona [10]. The main limitation of this technique has been that it was confined to the study of relatively large single crystals of a few mm in size at least. This was dictated by the size of the primary beam, which is typically of the order of one millimeter in diameter. Many materials of technological importance, however, are polycrystalline with much smaller crystallite sizes. Even on large single crystals a good lateral resolution is highly desirable in order to resolve domain structures, terraces and steps or a laterally inhomogeneous adsorbate coverage, to name only a few examples. Recently, an analytical reflection and emission UHV surface electron microscope has been presented [14] which, among other techniques, provides low energy electron reflection micrographs in the imaging mode.

In this paper we report on the development of an alternative microscopic LEED technique which we call "Scanning LEED Microscopy".

Basic considerations

Several approaches have been developed for surface imaging in electron microscopes besides the one mentioned above: (1) Reflection High Energy Electron Microscopy (RHEEM) [4,6,7,11,16], Scanning Electron Microscopy (SEM) with various types of signals and detectors [9,15], Photo-electron Emission Microscopy (PEEM) [2,3,12],
and mirror electron microscopy [1,5]. Not in all of these approaches had the requirement of surface cleanliness and ultra high vacuum been fulfilled, which are mandatory, however, to obtain meaningful results. A common feature of all approaches is to gain surface sensitivity by reducing the electron momentum normal to the surface and/or to select electrons with small energy loss or low energy for the image formation. In our approach the surface sensitivity is similarly obtained by using primary electrons of low energy and by discriminating the detected electrons against those with large energy loss. We use primary beam energies between 200 and 500 eV and a typical angle of incidence of 45° with respect to the surface plane. Under these conditions the electron momentum normal to the surface is very similar to that in conventional LEED, where electrons of around 100 eV energy impinge normally onto the crystal. In comparison to RHEEM the angle of incidence is very large, so that the foreshortening is quite acceptable and the requirements on the macroscopic flatness of the surface are much relaxed.

The basic concept of scanning LEED microscopy comprises two steps: in the first step a fine beam of low energy electrons is produced, and the resulting LEED pattern is recorded. In the second step the information in the LEED pattern is used to generate an image of the surface while the primary beam is scanned across the surface. As is well known, it is difficult to form intense electron beams at low energy. Because of the fundamental Liouville theorem it is necessary to start with a cathode of high brightness. Since the chromatic aberration of the lenses becomes a limiting factor at low beam energy, the energetic width of the emitted cathode must be small. At the present time both requirements together are best fulfilled by a field emission cathode. The primary beam currents are at least three orders of magnitude smaller than in conventional LEED (typically 1 pA). However, even if it were possible to extract similar beam currents, this would not be desirable. At a beam size of 0.1 µm the current density would increase by 6 to 7 orders of magnitude. Even leaving aside thermal effects (which would not be negligible), electron beam damage such as desorption and bond breaking would render such an instrument useless. This is, by the way, also a problem in conventional, high energy transmission or reflection microscopy. In order to keep the current density at the level used in conventional LEED, primary beam currents in the range of 0.1 to 1 pA are required. At these low levels observation with a conventional LEED screen is impossible. Even image intensifiers with a gain of 10^4 would not be adequate. Instead, one has to employ single electron detection. Doing this with a single electron multiplier would be cumbersome, though possible. We used the approach first tested by Stair [13] who placed a two-stage channel-plate multiplier behind the LEED retarding field analyzer and used a position-sensitive two-dimensional readout with a resistive anode. In this way each single electron diffraction event is observed as well as the particular beam into which the electron was diffracted. The corresponding LEED pattern can be observed and stored on an oscilloscope screen. By placing the primary beam at different locations on the sample, differences in the two-dimensional periodicity on the surface can be analyzed locally. This is shown schematically in Fig. 1.

Instead of taking LEED patterns point by point, alternatively one may select one (or more) of the diffracted beams and use its intensity to modulate the brightness of the display tube while the primary beam is rastered across the sample. In the simplest case, to be demonstrated below, one may select the specularly reflected beam and will obtain a 'dark-field image' of the surface. The lateral displacement of the LEE0 pattern as a whole during scanning corresponds to the extension of the field of view and will thus be negligible in most cases. There is a variety of other modes of image formation, which have not yet been fully explored. For example, if a thin adsorbate or metal layer exists in islands on the surface, causing a reconstruction or forming a superlattice by itself, these islands can be made visible by selecting the 'superstructure beams' in the LEED pattern for the intensity modulation. Then the display screen will appear light when the primary beam is on the island and dark, when it is on the substrate.

The experimental realization of the concept of Scanning LEED Microscopy is discussed in the next section.

Experimental aspects

The experimental studies have been made in the UHV-field-emission scanning electron microscope (SEM) described previously [8]. Fig. 2 shows an outline of the ultra-high-vacuum (UHV)-field emission SEM system having two chambers with vacuum better than 5.10^{-10} mbar. The right hand chamber is equipped with standard (microscopic) surface physics tools such as a conventional LEED system, a sputter ion gun, a quadrupole mass spectrometer, a deposition source and gas inlet. The left hand chamber contains the field emission microscope, a movable cylindrical mirror analyzer for Auger microanalysis and the channelplate LEED detector. The electron optical system and its control unit and display circuits are modified versions of the Hitachi S-800 microscope. Samples are inserted into a preevacuation chamber and transferred to the manipulators in the two UHV chambers by a magnetically coupled transfer rod. The electron optical column is composed of the usual electrostatic lens of the Butler type and two magnetic lenses. The electron beam is decelerated by the Butler lens when the final beam energy is lower than the extraction voltage. The whole optical system is shielded by µ-metal against stray magnetic fields. A beam size of 60 nm is obtained at 250 eV and of 2nm at 30 keV. The resolution and optimum magnification in the low energy regime is shown in Fig. 3 [8]. Even at 100 eV a beam size of less than 200 nm was obtained as judged from the imaging of gold
Scanning LEED Microscopy

field-emission microscope column

grid

channel plate

position-sensitive readout

deflection plates

scope screen

raster image

sample

Fig. 1

The basic concept of a Scanning LEED Microscope. For explanations see text.

coated polystyrol spheres of ~ 1 μm size. Preliminary work indicates that at low energies the resolution might be improved substantially by reducing the extraction voltage. The LEED detector assembly (see Fig. 2) consists of two hemispherical grids for energy analysis, a two-stage channelplate amplifier and a resistive anode, followed by a position sensitive detector (Model 239G, Surface Science Lab., Palo Alto). The detector assembly moves on circular rails around the sample, with a radius of approximately 60 mm. The angle of incidence of the primary beam may be chosen between grazing and about 60° with respect to the surface plane. The spatial resolution of the position sensitive detector is nominally 200 lines/40 mm, corresponding to an angular resolution of about 3 mrad. The convergence angle of the primary beam is confined to 1 mrad or less by a variable objective aperture. Thus, the 'transfer width' (or inverse momentum resolution) of the system is presently determined by the detector resolution. The x-y-coordinates of each electron diffraction event are given as two DC voltages. These are applied to the x-y deflection plates of a storage oscilloscope, together with a brightness pulse. If a scanning image in the dark field mode is to be formed by one of the diffraction spots, an electronic window is set

Fig. 2

Outline of the Ultra-high-vacuum field emission scanning electron microscope system. The microscope chamber (left) and the preparation chamber (right) are connected by a sample transfer device.

Fig. 3

Resolution (left-hand scale) and optimum magnification (right-hand scale) for low accelerating voltage. A resolution of about 60 nm is achieved at 250 eV primary energy.

Fig. 4

Schematic electronic circuit for setting an electronic window around a diffraction spot on the LEED detector.
around this spot. In its simplest form it is provided by two analog comparators (see Fig. 4). When the x- and y-coordinates lie within the limits, an event pulse from the position computing electronics is passed through the digital switch and produces a bright spot on the display tube of the electron microscope. This scheme may be extended in a straightforward way to include a group of diffraction spots or to form two different images from two diffraction spots in one run.

Results

As a first example Fig. 5 shows a micro-LEED diffraction pattern from a clean Si(111) surface. This pattern has been taken at 250 eV energy and a primary beam current of about 10^{-13} A. It was photographed from an oscilloscope screen with ~3 min integration time. We recall that this pattern has been taken via single electron counting, with about 10^6 electrons detected. The pattern stems from a spot about 100 nm in size located on a well-ordered reconstructed (111) terrace of a stepped Si(111) surface. The pattern is distorted relative to conventional LEED patterns because of the non-normal incidence of the primary beam. The encircled spot corresponds to the (00) beam ( specular reflection), while the majority of the spots belongs to the well known (7×7) superstructure. The rightmost spot along a horizontal line starting from the (00) beam corresponds to the (10) beam. This example shows that it is possible to resolve even highly structured diffraction patterns at relatively high energy.

The second example refers to the scanning mode, with polycrystalline Si as a sample. Fig. 6a shows a conventional SEM picture of the surface. In Fig. 6b a microdiffraction pattern is shown, as taken at the large flat structure at the center of the right half of Fig. 6a. The diffraction pattern shows an unreconstructed (1×1) surface at this location. The electronic window is set around the (00) diffraction spot as indicated. In the scanning LEED mode the intensity of this spot is used to modulate the brightness of the SEM display while the primary beam is scanned. The result is shown in Fig. 6c. The flat area of Fig. 6a appears bright, as well as some other spots near the upper edge of the frame. The absence of intensity in other parts of Fig. 6c means that the position of the specular beam in the LEED screen is outside the electronic window. Microdiffraction patterns from other locations, e.g. the two bright spots near the vertical center line in 6a), show also a (1×1) surface but with a different spatial orientation, so that the specular beam moves out of the window. This example shows that by scanning LEED microscopy polycrystalline samples become accessible to surface structure studies.

The final example demonstrates the application of scanning LEED microscopy to surfaces with thin layers of adsorbates. Fig. 7a shows the SEM picture of a Si(111) surface with an overlayer of Au. On this surface two kinds of characteristic micro-LEED patterns were observed. Fig. 7c shows the (5×1) superstructure, while Fig. 7d shows the (√3 × √3) superstructure. The former corresponds to a coverage of about 1/4 monolayer Au, while the latter one indicates a coverage of about 1 monolayer. A conventional wide-beam LEED pattern shows an intensity superposition of the two patterns. The scanning LEED micrograph in Fig. 7b indicates, however, that these two superstructures are distributed unevenly over the surface. Fig. 7b was taken with the specular beam intensity, showing a bright band near the center of the frame (the dark spots are holes in the surface as determined from an angular dependence of the contrast in SEM pictures). Unlike in the previous example the contrast does not arise because the specular beam moves out of the window. Rather, it comes out by a different (00) beam intensity in the (5×1) and (√3 × √3) superstructures. Because of multiple scattering of the electrons in this case the intensity is lower for the higher Au coverage (√3 × √3) than for the lower coverage (5×1). The diffraction pattern of Fig. 7c was taken at the bright area, that of 7d) from the darker area surrounding it. The bright band in Fig. 7b thus corresponds to a coverage of 1/4 monolayer of Au, surrounded by areas with about 1 monolayer Au. This example shows the usefulness of scanning LEED microscopy for the study of the lateral distribution of overlayers with monolayer or sub-monolayer thickness. We may mention that an elemental micro-analysis of the surface may be carried out with the Auger-facilities in the UHV chamber, with a resolution of about 30 nm.

Conclusions

The basic concept of Scanning LEED Microscopy has been outlined and its experimental realization has been demonstrated. The technique is complementary to existing techniques of surface imaging with perhaps the closest relationship to the analytical microscope of Telleps and Bauer [14]. The relationship between this approach and ours is the same as that between an imaging microscope and a scanning microscope. While they presently have a superior lateral resolution in the imaging mode (20 nm have been reported in ref. [14]), the resolution in the microdiffraction mode is less favourable (~10 μm spot size; anticipated improvement to <100 nm [14]). The main advantages of the Scanning LEED microscope are that the sample is in a field-free region during observation, that the sample surface is easily accessible during observation, and that samples of irregular shape pose no problems. The possible applications of Scanning LEED Microscopy are numerous and have not yet been fully explored.

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Micro-diffraction pattern of a clean Si(111) (7x7) reconstructed surface as obtained on an oscilloscope screen.

Fig. 5

Micro-diffraction pattern of a clean Si(111) (7x7) reconstructed surface as obtained on an oscilloscope screen.

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Fig. 6

a) SEM micrograph of a polycrystalline Si surface.
b) Micro-diffraction pattern taken on the large grain in a) showing a (1x1) pattern. The (0,0) beam is encircled.
c) Scanning LEED micrograph of the area in a) obtained by using the intensity in the (0,0) beam for brightness modulation.

c) Micro-diffraction pattern from the bright area in b), showing a (5x1) superstructure, due to a Au coverage of about 1/4 monolayer.
d) Micro-diffraction pattern from the dark lower left corner in b), showing a (sqrt(3) x sqrt(3)) superstructure with about 1 monolayer Au coverage.

Fig. 7

a) SEM micrograph of a Si(111) surface with a thin Au overlayer of varying thickness.
b) Scanning LEED micrograph as obtained by the intensity of the specular beam, as indicated in c) and d).
c) Micro-diffraction pattern from the bright area in b), showing a (5x1) superstructure, due to a Au coverage of about 1/4 monolayer.
d) Micro-diffraction pattern from the dark lower left corner in b), showing a (sqrt(3) x sqrt(3)) superstructure with about 1 monolayer Au coverage.
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J.M. Cowley: Values are quoted for the "beam resolution" of the instrument for various voltages. How is this resolution defined and how was it measured?

Authors: The word "beam resolution" refers to the inherent resolution limited by the size of the electron beam at the sample, disregarding all other factors influencing the resolution adversely (in our case mainly sample stage instabilities). It was determined from SEM pictures of test samples under conditions of minimal vibrations as described in ref. [8].

J.M. Cowley: The magnification of the figures 6c) and 7b) are not sufficient to display the resolution achieved. What resolution could be obtained for such cases and what factors limited the resolution? What accelerating voltage was used in these cases?

Authors: The maximum possible resolution is given by Fig. 3. In the present case the resolution was mainly determined by sample vibrations and is certainly inferior. The obtainable resolution in the scanning LEED micrographs is about the same as in SEM pictures, disregarding sample vibrations. The accelerating voltage was 500 V in Fig. 6c) and 250 V in Fig. 7b).

J.M. Cowley: It is known that channel-plate amplifiers are limited by the large time constant for individual channels. How many channels were used to detect the signals for the images, (figures 6c) and 7b)), and what was the recording time for these figures?

Authors: The number of channels involved in the detection of one beam (one was used in the figures) is of order 10^4. Thus, the dead time of an individual channel is not the limiting factor. Rather, it is the electronic dead time of the position sensitive detector. The recording time for the figures was about 10 min.

J. Venables: Can you say more about the maximum count rate of your position sensitive detector? The problem seems to be that you have to reduce the primary beam current to ~10^-13 A (Fig. 5) to come within this counting limit, whereas with a TV or CCD detector more reasonable primary currents (10^-11 - 10^-12 A) could be used.

Authors: It is true that the primary beam current had to be reduced to stay within the count rate limit. With the present electronics good quality micro-diffraction patterns can be recorded with up to 2x10^6 cps. The resolution degrades at higher count rates until at 10^7 cps the dead time limit is reached. In the most recent devices this has been extended to above 10^6 cps.

We think that in very much of the surface work to be done with Scanning LEED microscopy one will prefer to stay in the current range of one to perhaps several tens of pA and to use single electron counting. The reason is that the beam induced damage at the surface, in particular with gaseous adsorbates, puts severe limitations on the electron dose applied to the sample. If we assume a current of 1 nA into a spot of 5 nm and a measurement time of 100 sec, this corresponds to a dose of about 5x10^3 As/cm^2. Even with a rather stable compound like oxidized aluminum this would lead to almost complete disintegration according to measurements of Pantano and Madey (Pantano C G and Madey T E, Appl. Surf. Science 2 (1981) 115) taken at 5 keV beam energy. Even if we assume the damage to be ten times less at 500 eV (which is overoptimistic), a very substantial fraction of the oxygen would be removed during the measurement. Since CCD and TV devices need relatively high currents, their use will probably be restricted to the study of very stable systems.

J. Venables: Can you say what probe current is available as a function of resolution at low probe energy?

Authors: These numbers have not been measured over the whole range of energies. As a representative value a current of 2x10^-10 A at 200 eV with a 'beam resolution' of 100 nm was obtained (see ref. [8]).