# Low energy electron diffraction (LEED) on single crystal metal surfaces

Manual for the laboratory course NEVF132
Fall 2012

In this laboratory students will get acquainted with the method and instrumentation for the static and dynamical low energy electron diffraction (LEED). Part of the ultra-high vacuum (UHV) instrumentation may be already familiar to them thanks to the other experiments in this laboratory course. Of the dynamic LEED method students will do the experimental part and data processing into the I-V curves only, not the complementary tensor LEED modeling. This text focuses on the practical laboratory work; it does not by any means supply the necessary amount of theoretical knowledge required. Theory should be sought in and learned from the cited references [1-3] and other articles and monographs, many of which are cited in [2].

## Introduction

Diffraction is a phenomenon first described in optics and scattering of light (photons) off optical apertures, grids and other elements. Diffraction patterns are sensitive to characteristic dimensions of the scattering elements, so that evaluation of the patterns can be used to determine those dimensions. This sensitivity is largest when the characteristic size measured is similar to the wave length of the probing photons. Microscopic objects are thus well measured in the optical range of photons, interatomic distances in solids are revealed using x-rays.

Scattering experiments with electron beams incident on metal single crystals (the experiments on nickel by C. J. Davisson and L. H. Germer in 1927 being the first [4]) confirmed the wave character of electrons and showed the use of electrons to probe the crystal structure of matter. Kinetic energy of electrons translates to wave length, which is as important a parameter of diffraction as photon wave length in the scattering of light. In the formula

$$\lambda = \frac{h}{\sqrt{2m_0 eU}} = \frac{1{,}23\cdot 10^{-9}[\sqrt{V}m]}{\sqrt{U[V]}} = \sqrt{\frac{150\,[V\cdot Å^2]}{U[V]}} \quad , \qquad (1)$$

the $U$ is the accelerating voltage of the electron beam source and then $eU$ equals the kinetic energy of the electrons. As the interatomic distances in solids are in the order of $10^{-10}$ m, the electron energies from 1 eV up to several keV are suitable for studying the solid crystal structures.

A three-dimensional array (grid) of atoms in a single crystal then produces a pattern with bright spots on a phosphorescent screen of a standard LEED instrument. The spots appear (or rather are the brightest) at certain angles $\theta$ and natural ($n$) multiples of electron wave length $\lambda$. The Bragg's law, which ties these angles with the crystal lattice plane spacing $d$ through $\lambda$, may be reformulated thus:

$$d = \frac{n\lambda}{\sin\theta} \quad . \qquad (2)$$

The interatomic distance can be derived from $d$ upon the knowledge or assumption of the surface crystal plane orientation. As far as the elevation angle $\theta$ is concerned, the Bragg formula is equivalent to the Ewald's sphere selection rule [5] and Laue's equations [6] used in most chapters about LEED such as [2]. In order to understand the azimuthal angle $\varphi$ as well, one needs to get acquainted with the Fourier transform and reciprocal space metrics, which form the mathematical foundation of the kinematic theory of LEED.

A beam of monochromatic electrons colliding with the surface of a solid can be viewed as a plane wave. In real conditions this is achieved by collimation of the beam to minimize its divergence. As the electrons scatter elastically off the individual atoms of the material, the atoms act as individual sources of spherical waves following the Huygens-Fresnel principle. The constructive interference between these waves produces beams of scattered electrons emanating from the material in the directions given by Bragg's law.

Unlike X-ray light beam, the incident beam of electrons beam penetrates not the whole physical sample but reaches only to a certain mean depth in any given material. This is due to both the elastic and inelastic scattering. Several percent of the beam are removed at every atomic layer and the elastically scattered electrons get attenuated on their way back to the surface as well (further inelastic scattering) so that the typical information depth of a LEED pattern is a dozen atomic layers or so. This effect makes the LEED very surface sensitive. You can find graphs of the relevant inelastic mean free path (IMFP) of electrons in literature.

The trade-off, compared to the x-ray diffraction, is that LEED is limited to electrically conductive crystals only. The electron beam delivers the charge at the rate of ~ 0.1-10 μA, which cannot disperse fast enough in insulators and undoped semiconductors. The charge builds up in the illuminated part of material and distorts the scattered beams too much to be of use. The only way to measure insulators is to measure them in the form of very thin films only a few atomic layers thick, deposited on well-chosen conductive substrates. This may add complexities to the studied system, however.

The LEED instrument is used for standard scientific work as well as for this laboratory course. If machine maintenance or work traffic interferes with your planned lab time, your instructor may re-schedule your lab time. He/she will endeavor to announce the changes well in advance and set the new time at the earliest convenience of all.

## Laboratory tasks

1. A proper single crystal sample will be installed for your experiment in the UHV chamber. This will be a single crystal of copper, vanadium, or palladium. Before the lab time, ask the instructor for any details about it that you consider important for the laboratory experiment or data analysis.
2. Whether you are going to make a laboratory report from this experiment, make notes on the experiment as if the report was planned. You will leave a copy of your notes with the instructor after you finish the experiment.
3. Practice fixing a sample onto a sample transfer plate. If working in a group, one student does this while the rest clean the sample.
4. Clean the sample surface by $Ar^+$ bombardment and subsequent heat annealing until acceptable diffraction pattern is produced.
5. Produce a focused static LEED pattern on the luminescent screen, refine the sample alignment and take snapshots of the pattern. Your report must contain lattice parameter values calculated from here.
6. Try to modify the surface using a method suitable to your sample. Take snapshots of the resulting LEED pattern. Evaluate the pattern in your report.
7. Choose a sensible energy range and take a dynamic LEED measurement – series of snapshots with about 1 eV energy increments. Extract the I-V curves from this series for your report.
8. Discuss the I-V curves qualitatively.

## Checkpoint

1. How does thermocouple work? What is the thermocouple reading, when sample is at 21°C?
2. What is the highest background pressure (order of magnitude) such that LEED works?
3. What is the exposure (rate of incoming molecules) of the sample surface to dirt from the residual gas in the chamber at a pressure $10^{-7}$ Pa?
4. What will be the difference between LEED patterns of Cu(111) and V(110) at energies, where each give the brightest spots?
5. What do we expect to observe in the LEED pattern of a good crystalline surface as we change the beam energy from 0eV up?
6. How do we make an I-V curve out of the series of snapshots?
7. What are the differences between LEED, XRD and RHEED methods?
8. How will a band gap (the gap around Fermi energy) in the electronic density of states affect the LEED results?

## Laboratory report requirements

1. Limit the "Introduction" section to an overview and to the facts really needed to understand the laboratory and analyze the data. Use your judgment.

2. Likewise, limit the description of instrumentation to a diagram with the key components and the principles of function. No need to copy the operating manuals, but publish the exact step-by-step operating procedures and point out, if and how they differed from those in this manual.
3. Do **not** add the answers to the "Checkpoint" quiz to your report; they are for lab time evaluation only.
4. Focus on describing and explaining the data processing and analysis you did.
5. Be sure to perform all the analysis and publish the results specified in "Laboratory tasks" section.
6. Make your graphics a good and concise documentation of your work and the data analysis. Graphs usually do better than tables.
7. Estimate errors of measurements where possible. You are welcome to ask any questions regarding the instrument and technique, which remain unanswered in the references named at the end of this document.
8. Be prepared to submit your report in electronic format to the LEED lab instructor, who will do the grading. Make your report sensible in all senses; consider that even unnecessary waste of ink/toner (large black areas in static LEED images) or electronic documents too large may negatively affect your grade.
9. A folder with your **raw** data and the **I-V curves** data file (e.g. spread sheet document) must be available to the instructor at the time of grading.
10. You need not show all the raw data in the report. In fact, plain duplication of raw data in the report is strongly discouraged!

## Instrumentation

### *Vacuum chamber*

The LEED instrument and its auxiliary appliances are placed in a standard stainless steel vacuum chamber. The operating vacuum is usually $1 \cdot 10^{-7}$ Pa or better, except during sample surface cleaning and preparation. All parts facing the vacuum are made of low vapor pressure materials. There is no load lock at this chamber, so after each sample exchange the chamber and all its components need to be outgassed for correct function. This is a lengthy process, which is why the assistant does it for you before your lab time.

### *LEED optics and its controller*

The LEED instrument present is a 3-grid model of the ErLEED-150 operated with the ErLEED 1000A analog power supply. Refer to documents [11-13] for parameters of the instrument and operation instructions.

### *LEED camera and snapshot recording*

The luminescent screen is filmed by a CCTV camera, which sends the analog TV signal to a video card in a local computer. Software is installed on the PC that allows viewing the TV signal and

record snapshots in standard image formats or in a binary format. These snapshots will be your raw data.

### *Vacuum pumps*

The system is kept at high or ultra-high vacuum by Pfeiffer turbomolecular pumps (TMPs) backed by an oil-free scroll pump. The main chamber pumping line must be up and running before, during and after your experiment, so you need not concern yourself with their operation. If the TMP on the ion beam source is stopped, when you start your lab time, you will need to start it and let it run at full speed for a while. Each TMP is switched on and off via the front panel of its own controller.

### *Vacuum gauge*

A cold cathode vacuum gauge is installed at the main chamber. It starts to give readings below $1 \cdot 10^{-1}$ Pa. It should be running for the sample cleaning, but disconnected for the LEED measurements themselves. This (dis)assembly ought to be done under supervision.

### *Manipulator*

A 5-axis manipulator is used for the sample alignment needed in dynamic LEED measurements. **Note: Be advised that pushing the limits of sample position and orientation may lead to mechanical damage, so don't push it!**

The movement ranges are 10 mm in the horizontal plane (x-axis parallel to the LEED axis, y-axis perpendicular to it) and 55 mm along the vertical direction (z-axis).
The angle of rotation about the z-axis is denoted η and is limited to ± 40° from the orientation of normal incidence of the electron beam.
**Note: The η rotation limits are set by collision of the manipulator body with the LEED shutter. Use visual control (through window or via LEED pattern) every time you change η!**

The tilt, i.e. the sample rotation about a horizontal axis parallel with the sample surface, is denoted θ and is limited to about ± 10° by manipulator design. It has no other function than to fine-tune the alignment of the sample surface normal with the LEED axis.
**Note: The tilt is a very FRAGILE part of the current setup, don't regulate it unless instructed to do so! It is FORBIDDEN to move tilt from its pre-set value (record before manipulation) by more than 2 mm in either direction!**

The e-beam source is currently not perfectly parallel with the rotational axis of the LEED grids and screen, so the "normal incidence" of electrons is achieved at approx. 3,3° - 4,3°, i.e. 6,0-6,3 mm off the manipulator tilt scale. As the sample transfer plate and manipulator align the sample fairly well, tilting the sample by more than 1 mm, i.e. about 3-4° in θ, is NOT necessary.

### *Sample transfer plate*

The transfer plate design depicted in Fig.1 shares the general layout of interface with several other instruments in the Surface Physics Laboratory, namely the STM (Dr. Mysliveček) and the ESCA (doc. Dr. Veltruská). The details of sample fixation, thermocouple position and heating implementation vary among these machines. You will get acquainted with the details of our design during the lab-time practice assembly.

In our particular case, the sample is positioned as a lid on top of a molybdenum box (a "furnace") that contains a coiled tantalum filament for sample heating. A chromel-alumel thermocouple is affixed to the furnace. This block – the thermocouple with its contacts, the furnace, the sample, and the tantalum foil that presses the sample to the furnace – are electrically interconnected, but isolated from all the other parts.

Figure 1: The sample transfer plate.

The tantalum filament mentioned above is fixed to its contacts on the two sides of the transfer plate and electrically isolated from everything else. Finally, the body of the transfer plate is made of nonmagnetic stainless steel or molybdenum. During measurements it is mechanically and electrically connected to the manipulator, which is grounded. Notice that this does not provide grounding to the sample. It, nevertheless, avoids charging, which would produce an electrical field that would affect the electron trajectories and ruin the measurement.

### *Ion source and its power unit*

Refer to the manuals [7] and [8] for the description and operating instructions. The ion source can be operated in a static mode or scanning mode, you will only use the static mode with minimum focus. Remember this and don't lose time with the scan-specific controls. **Operate the ion source and its power unit under supervision only. When in doubt, ask!**

## Procedures

### *Sample alignment*

There are three principal alignment positions in the system; two of them are to be used in this experiment. The x, y, z and rotation η values are given here according to the dials on the manipulator knobs. The tilt θ angle is to be set approximately by visual means, dial value is only approximate.

**Note: Make sure the rotation η is set near 0° before the vertical linear (z-axis) motion. Damage to instrument may result otherwise!**

<u>sample cleaning by $Ar^+$ sputtering:</u>

x = 16 mm
y = 21 mm
z = 42.5 mm
θ = the LEED value for tilt works fine
η = 30° for actual sputtering;

**Note: Set η = 330° before starting the $Ar^+$ ion gun and use the rotation to 30°and back to start and stop the sputtering run.** Sputtering works best at an angle ~45° off sample surface normal, which is fairly taken care of by the 55° inclination of the ion gun flange.

<u>LEED measurement:</u>

x = 16-27 mm

**Note: The position of sample should be 23 mm from the μ-metal shield edge, which corresponds approximately to x = 21 mm. The instructor may instruct you to use another value, however.**

y = 16-21 mm
z = 16-25 mm

**Note: Choose z position according to the quality of the LEED reflections.**

θ = 3,3-4,3° … i.e., tilt dial ~ 6-6,3 mm (could be different)
η = 0°

Note: Both angles must be fine-adjusted by direct visual control (through window) and with LEED pattern observation. In the latter case, the **patterns must be symmetrical and spots of equivalent reflections must brighten and dim simultaneously during beam energy change**.

### *Sample cleaning*

<u>Ar+ ion bombardment</u>

1. If the turbomolecular pump (TMP) of the differential pumping line is off, turn it on.
2. Check if the vacuum gauge is connected to instrument and turned on.
3. Check the status of the differential pumping valve (DPV) and open it fully if not yet open.
4. Check (ask instructor) the status of the ion source interconnecting valve (ISIV) and if closed, open it by 2/3 of a turn.

5. Check (ask instructor) the status of the Argon tube. It can be (a) under vacuum, (b) filled with air, or (c) filled with pure Argon. In cases (a) and (c) proceed by opening the Argon cylinder; in case of (b) ask the instructor for advice.
6. Ground the sample.
7. Make sure that the LEED optics shutter is well closed.
8. Set the manipulator to the sample cleaning position, set η to value prescribed for ion gun start-up.
9. Observe the pressure closely and start the ion source power unit.
10. Open the argon cylinder.
11. Open the gas inlet valve (GIV) slowly and regulate chamber pressure to ~ $1 \cdot 10^{-4}$ Pa.
12. Following the manuals [7] and [8] run the ion source for the **High Beam mode** (i.e. with Focus2 = 0) at 1 kV and emission current 10 mA (or other settings given by instructor).
13. Rotate the sample to the cleaning position and let it be sputtered for the desired time. Then stop the sample sputtering by rotating away from ion source.
14. Turn the ion source power unit to Standby and close the GIV. Close the argon cylinder.

Should you need another sputtering cycle, you may repeat this procedure starting at **step 6**, because you leave the differential pumping on and the gas line filled with pure argon.

Annealing

15. Connect a voltmeter to the thermocouple feedthrough and a DC power supply to the heater feedthrough.
16. Check again that the LEED optics shutter is shut.
17. Heat the sample to the annealing temperature appropriate to the material.
18. Anneal for a few seconds, then turn the power supply down and off and allow the sample to cool down. You may start the LEED power-up procedure, but wait for the sample temperature to drop below 100°C before a LEED measurement.

### *LEED measurement*

A successful LEED measurement relies upon an outgassed filament of the electron gun. This filament activation takes several hours and must be done at least once after insertion of every sample. It is your instructor's responsibility to arrange this.

There are two applications installed on the LEED PC to record the LEED data, but the instructions "take a snapshot" below may refer to either one of them. Firstly, there is the application that belongs to the Leadtek WinFast framegrabber card. This records the camera signal as standard monochrome (greyscale) images with 8-bit information depth. Secondly, the software AIDA from the LEED manufacturer can do the same job, except it saves the images in a binary format. This format cannot be directly visualized by general image viewers, but offers up to 16-bit depth. This offers a much better signal-to-noise ratio, so good data is not degraded in processing. Depending on the sample and the LEED pattern quality your instructor will choose

the suitable method of data acquisition. Naturally, he/she will then advise you on how to load the data to some scientific software for analysis.

1. Check the chamber pressure.
2. Check that LEED optics shutter is closed, sample is grounded and manipulator in the approximate position for a LEED measurement.
3. Turn off the vacuum gauge sensor. Disconnect and remove the vacuum gauge body and magnet and store all in an area appointed by the instructor.
4. Power up the LEED power supply, the CCTV camera and the computer.
5. Following the manual [11] section 5.2 and test report [12] adjust the offsets and gains of the electron gun (beam source) voltages: Wehnelt ($U_W$), Anode ($U_A$), Lens 1/3 and Lens 2. You only need to check and adjust settings for E=0V and E=300V.
6. Now activate the filament. The long activation has already been done, so only a short one is needed. Set E = 500 V, $U_W$ = 0 V and $U_A$ = 500 V. Increase the filament current $I_C$ at a moderate speed up to 2 A, or until you observe a positive anode current $I_A$. Then slowly raise the $I_C$ up to 2.55 A. Wait until the anode current reaches 0.4 mA, then lower the $I_C$ to 2.35 A.
7. While waiting for filament activation, run the framegrabbing software.
8. When filament is ready, set the Suppressor voltage $U_{supp}$ to maximum, set Screen voltage $U_{screen}$ to 6 kV and open the shutter. A LEED pattern should appear.
9. Adjust and optimize the sample position and orientation. **Be extremely careful with the tilt adjustment!**
10. Adjust the $U_W$, $U_{supp}$ and $U_{screen}$ voltages so that the spots are not overly bright. This means that in the Preview window of AIDA the brightest pixel has intensity less than 100%. A fairly good value is 90%. A saturation of the CCD camera chip would ruin your I-V curves measurement.
11. Try to identify the diffraction spots as they appear on the screen and take snapshots at some suitable energy values.
12. Choose a suitable energy range over which the spot intensities change noticeably. Fine-tune the sample alignment so that equivalent reflections maximize their brightness at the same energy. **Since intensity increases with beam energy E, re-adjust the intensity of reflections (like in step 10) near the maximum of the chosen energy interval.**
13. Take a series of snapshots along the chosen energy interval with 1 V or 2 V steps. Make sure you record the primary electron beam current at each step – it is used in analysis for I-V curves' normalization.
14. After data acquisition is finished, close the LEED shutter, set $U_{screen}$ =0 V, set E = 0 V, turn $I_C$ down slowly until $I_C$ = 0.0 A, then turn $I_C$ off.
15. Quit the data acquisition application, unplug the camera; check that ion beam source power unit is off, the argon cylinder is shut; the heater power supply is off, the thermocouple voltmeter is off. **Leave the pumps running.**

### *LEED data analysis*

There are two types of analysis you are going to apply. Firstly, you will make a geometrical analysis of an image (or a few images) taken at some energy and from the positions of the bright spots will deduce the atomic plane distance and the lattice constant of the material. For image size calibration you will need to measure the physical dimensions of the LEED screen and observe the parameters of the spherical geometry.

Secondly, you will need to extract the intensities of the bright spots for a series of images. You then plot these intensities as functions of beam energy. You obtain the intensity of a spot by first subtracting the background and then integrating the rest. Observe that the quantity "Intensity" thus obtained is not measured in anything near the real physical units, i.e. the rate of electrons per steradian. The measured quantity is only (nearly) proportional to the real intensity. A calibration is not needed, only normalization to the primary beam current is.

For the intensity extraction (building I-V curves) you are encouraged to use any scientific software you are comfortable with, open source applications are as good as commercial ones. The University, the Faculty and the Surface and Plasma Physics Department supply you with a wide range of commercial programs, a list of which is available at [14].

## Bibliography


[1] **M. A. van Hove**, "Low-Energy Electron Diffraction: Experiment, Theory and Surface Structure Determination" in "Springer Series in Surface Sciences", Springer 1986.

[2] **J. Komrska, F. Máca, and M. Láznička**, chapters in "Metody analýzy povrchů", vol. "Elektronivá mikroskopie a difrakce", eds. L. Eckertová, L. Frank, Academia Praha 1996

[3] **N. W. Ashcroft, N. D. Mermin**, "Solid State Physics", 1st Ed., Brooks Cole 1976

[4] **C. J. Davisson, L. H. Germer**, articles dating 1927 through 1930

[5] **P. P. Ewald**, "X-ray diffraction by finite and imperfect crystal lattices", Proc. Phys. Soc. 52, 1940, p. 167

[6] **M. von Laue**, "The diffraction of an electron-wave at a single layer of atoms", Phys. Rev. 37, 1931, p. 53

[7] "Ion Source IQE 12/38 User's Manual", Specs Surface Nano Analysis GmbH, ver. 1.5, 1998

[8] "PU – IQE 12/38 Manual", Specs Surface Nano Analysis GmbH, ver. 1.4.2, 2000

[9] **C. J. Powell, A. Jablonski**, "NIST Electron Inelastic-Mean-Free-Path Database Users' Guide", ver. 1.2, 2010

[10] **S. Tanuma, C. J. Powell, D. R. Penn**, articles dating 1988 through 1994

[11] "Optics and Power Supplies ErLEED User Manual", Specs Surface Nano Analysis GmbH, ver. 1.3, 2003

[12] "LEED Optics Test Report: ErLEED s.n. 0366", Specs Surface Nano Analysis GmbH, 2003

[13] "ErLEED 100/150 – Reverse View LEED Optics" leaflet, Specs Surface Nano Analysis GmbH

[14] http://physics.mff.cuni.cz/kfpp/, menu "FAQ & kuchařky" > "SW licence"