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LOW ENERGY ELECTRON MICROSCOPY WITH ESCHER

We describe the layout and the capabilities of a new aberration-corrected low energy electron microscopy (LEEM) and photo electron emission microscopy (PEEM) facility, which features real- and reciprocal-space spectroscopy. This new setup, named Electronic, Structural, and Chemical Nanoimaging in Real Time (ESCHER), was recently installed at Leiden University. It has three major instrumentation-related goals. First, we aim to reach the ultimate spatial resolution facilitated by aberration correction using an electron mirror, together with advanced electron detection. Second, we want to develop and exploit the spectroscopic possibilities of LEEM and PEEM in a standard laboratory environment. To this end, ESCHER is equipped with an inline energy filter and advanced photon sources. Third, we plan to extend the sample temperature range down to approximately 10 K, which is significantly lower than that achieved to date. Combined, these efforts will broaden the scientific reach of LEEM and PEEM beyond the areas of surface and materials science and into the realms of biosciences and life sciences. Here, we also present images of the first experiments performed with ESCHER focused on the growth of graphene on SiC(0001).

2.1 INTRODUCTION

Low energy electron microscopy (LEEM) and photoemission electron microscopy (PEEM) are powerful surface-science techniques. They enable real-time in situ imaging of surfaces and interfaces at elevated temperatures and with nanometer resolution [1–4]. In this chapter, we describe an advanced aberration-corrected LEEM/PEEM facility, i.e., Electronic, Structural, and Chemical Nanoimaging in Real Time (ESCHER), which was recently installed at Leiden Center for Ultramicroscopy, Leiden University. The scientific goals of this Dutch national facility are threefold: 1) to reach the ultimate spatial resolution promised by electron-mirror-based aberration correction; 2) to develop, optimize, and exploit the spectroscopic possibilities afforded by inline, real-space, and reciprocal-space resolved electron energy spectroscopy; and 3) to extend the sample temperature range to cryogenic temperatures in the 10-K range or lower. In combination, these three goals will facilitate novel interdisciplinary experiments in scientific areas such as nanoelectronics, condensed matter physics, and biophysics. This will significantly broaden the scientific impact of LEEM/PEEM beyond the traditional areas of surface and materials science.

Here, we give a comprehensive description of the ESCHER facility and its current capabilities. We also present the first experimental results obtained during the installation and testing phase of the ESCHER instrument. Next, we present measurements of the chromatic and the spherical aberrations demonstrating successful correction of these aberrations. Furthermore, we present measurements of a record spatial resolution of 1.4 nm afforded by aberration-correction. Finally, we discuss the additional steps necessary to reach a spatial resolution close to the theoretical limit of about 0.5 nm (see chapter 3) with our instrument.

2.2 ESCHER SETUP

The ESCHER setup is based on the commercially available FE-LEEM P90 instrument (SPECS GmbH, Berlin) originally designed by IBM [5]. Figure 2.1a shows a schematic diagram of the low-temperature sample chamber design concept that we will discuss in detail in chapter 7. Figure 2.1b shows a photograph of the ESCHER setup. In the center, we see the vertical electron column that extends from the gun (top) to the image screen (bottom). Extending to the sides are the high-(right) and low-temperature (left) imaging chambers, plus their respective sample loadlock systems. In a high-temperature LEEM experiment, the electrons follow the optical axis indicated in purple and red in Fig. 2.1c. The cold field-emission gun generates an electron beam with an energy of 15 keV. A combination of a gun lens and a condenser lens serves to focus the electron beam with variable magni-
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fication. Next, a magnetic prism array, with electron optical properties that closely resemble those of a thin lens, deflects the electrons by 90° toward the objective lens and the sample. The sample itself is held at a negative potential close to that of the field emitter in the electron gun. Hence, the electrons can be decelerated to an energy in the range of 0 – 100 eV. After interaction with the sample, the electrons are reflected back into the vacuum and again accelerated to 15 keV in the direction of the magnetic prism array. The objective lens, plus a transfer lens, places a magnified image of the sample on the diagonal plane of the prism array. The prism array deflects the electrons again by 90° toward a second prism array. The midplane between these two prism arrays coincides with a diffraction plane. In a LEEM experiment, this is where the low-energy electron diffraction pattern is located. In a PEEM experiment, this is where one finds the angular distribution of the photoelectrons. An electrostatic lens located in this same plane transfers the image rotation-free from the diagonal of the first magnetic prism array to the diagonal plane of the second prism array. The latter deflects the electrons once more by 90° toward a four-element electron mirror. The purpose of this electron mirror is to compensate both the chromatic and spherical aberrations of the objective lens. This improves the image resolution from approximately 5 to less than 1 nm, as predicted by full wave-optical resolution calculations (see chapter 3). At the same time, the transmission of the instrument is increased by about a factor of 10 (see chapter 3). On the path toward the mirror, an uncorrected image is placed in the mirror object plane. The mirror reflects the image into that same plane, with a magnification of 1, removing chromatic and spherical aberration in the reflection process. The electrons then return to the second prism array, with the corrected image again located on the prism diagonal. After this final deflection of 90°, the electrons enter the projector system. By changing the settings of the projector column, either the real space image or the diffraction pattern can be projected onto the image screen.

In PEEM experiments, the emitted electrons are accelerated away from the sample and follow the same path as in a LEEM experiment. To generate photoelectrons, we can illuminate the sample using either a Hg discharge lamp or a focused He I/He II discharge source.

The base pressure in the ESCHER setup is about 10^{-10} mbar. Experiments can be performed at pressures up to approximately 10^{-5} mbar. Samples are cleaned in situ by heating via electron bombardment. Because images are obtained in real time, processes such as growth, interface formation, phase transitions, and phase transformations can be followed at high resolution and at video rate. The high-temperature imaging chamber is equipped with an evaporator source, as well as
Figure 2.1: ESCHER setup: (a) Schematic of the low-temperature sample chamber design concept; (b) photograph of the ESCHER setup; (c) schematic diagram of the ESCHER setup. The sample chamber for experiments at elevated temperatures (300 – 1800 K) is located on the right-hand side of the central gun/projector column. The cryogenic sample chamber (10 – 300 K) is placed on the left-hand side of the central column. The respective electron paths are indicated by the purple and blue (cryogenic temperature) and the purple and red (high temperature) lines. (HTS: high-temperature superconductor.)
with chemical vapor deposition facilities with a direct line of sight to the sample. A Hiden Hal 7 quadrupole mass spectrometer with a mass range of 500 u gives tight control over these processes.

One of the unique features of ESCHER is the implementation of two sample chambers positioned around a single vertical gun/projector column. This provides two experimental locations using only one set of central column components. In the right chamber, the sample temperature can be varied between 300 and 1800 K. The left chamber is equipped with a cryogenic stage and an objective lens to cool the sample to around 10 K (see chapter 7). By changing the polarity of the magnetic field in the prisms, we are able to switch between the high- and low-temperature imaging systems (see Fig. 2.1c). Note that ESCHER is equipped with two aberration-correcting mirrors. For the aberration correction to work, the 90° deflection angles provided by the magnetic prism arrays must always be of the same sense (either clockwise or counterclockwise) to maintain the proper symmetries in the imaging optics.

For distinguishing nanostructures, the highest possible spatial resolution, as well as the highest possible electron transmission, is desired. Clearly, electronic and mechanical stabilities are a first requirement for this. We minimize the effects of chromatic aberrations by using a cold field emitter with a small energy spread (0.25 eV). Even with the correction of the chromatic aberration coefficient $C_C$, higher rank aberrations make the choice of the electron source relevant. This is particularly important for experiments at electron energies at the sample below 5 eV, as preferred for imaging organic systems. Passive electromagnetic shielding of the entire electron path and active vibration isolation reduce external influences to a minimum.

To extend the possibilities even further, the ESCHER setup has been equipped with an inline energy filter [6]. This filter is based on the fact that the deflection angle of a magnetic prism is highly sensitive to the precise energy of the electrons. The current design provides an energy resolution of 150 – 200 meV at 15 keV electron energy ($\frac{\Delta E}{E} \approx 10^{-5}$), without the need for deceleration optics. In this configuration, the microscope can be used as a space- and angle-resolved ultraviolet photoemission spectroscopy facility for surface electronic structure studies. By combining information from both real and reciprocal spaces, spectroscopy can be locally carried out (microspectroscopy), or the spectroscopic information can be used to create contrast in real-space images (spectromicroscopy). Alternatively, when the sample is illuminated with the electron beam, the energy filter can be used to perform spatially resolved electron energy loss spectroscopy experiments.
2.3 **FIRST EXPERIMENTAL RESULTS**

Initial experiments performed with ESCHER during the installation phase concerned the growth of graphene on a 4H-SiC(0001) substrate (see, e.g., [6]). Graphene was formed in situ by heating the SiC substrate to about 1700 K [6]. Bringing the sample to these elevated temperatures causes the desorption of Si from the sample surface. Therefore, the SiC(0001) surface undergoes a sequence of surface phase transitions (see chapter 6) before a carbon-rich surface structure is formed. The carbon atoms left behind on the surface rearrange to form a thin graphene film with a thickness in the range of 1 − 4 atomic layers. Figure 2.2a shows step edges of the SiC substrate at an electron energy at the sample of $E_0 = 2.5$ eV. Figure 2.2b shows an image of the same area of the sample at a slightly higher electron energy of $E_0 = 4.5$ eV. We note the remarkable change in contrast. Hibino et al. [7] showed that the reflected intensity of a graphene/SiC(0001) sample in LEEM shows quantized oscillations as a function of electron energy (i.e. electron wavelength) and graphite thickness. The observed intensity peaks and dips can be understood in terms of interference between electron waves reflected from the sample surface and from the interface between graphite and substrate [7]. The change of contrast as a function of electron energy allows one to determine the number of monolayers of graphene at each position on the sample with atomic layer resolution [7]. Such an analysis reveals that the bright and dark areas in Fig. 2.2b correspond to one monolayer and two monolayers of graphene, respectively. Both images show a field of view of about 4.5 µm. For these images, the spatial resolution is limited not by the electron optics but by the spatial resolution of the multichannel plate.

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**Figure 2.2**: LEEM images of the same area of a SiC(0001) sample covered with a few monolayers of graphene taken with the ESCHER setup: (a) Step edges at the substrate are visible. The electron energy is $E_0 = 2.5$ eV. (b) The bright areas correspond to one monolayer, and the dark areas to two monolayers of graphene. The electron energy is $E_0 = 4.5$ eV. The field of view is approximately 4.5 µm.
2.4. **Aberration-corrected LEEM in Practice**

The goal is to correct third order spherical and lowest rank chromatic aberrations of the objective lens in LEEM in practice. To achieve this it is necessary to have precise control over the aberrations introduced by the aberration-corrector and also to quantify the total chromatic and spherical aberrations by measuring them. The measurement results can be used to re-adjust and improve the setting of the corrector if required. Finally, the error of the aberration measurements determines how good the aberrations can reliably be corrected.

First, we will introduce the electrostatic electron mirror and its optical properties. Then, we will outline ways to measure the aberrations $C_C$ and $C_3$ in LEEM. Successful aberration-correction in ESCHER is demonstrated by measurements of chromatic and spherical aberrations and by high resolution measurements of graphene on SiC(0001).

2.4.1 **Electron Mirror**

The ESCHER instrument is equipped with an electrostatic electron mirror capable of introducing aberrations with the same magnitude but opposite sign com-
pared to the objective lens. The mirror is placed in the electron optical path between the cathode objective lens and projector optics (see Fig. 2.1c). It consists of four elements, namely a ring-shaped electrode at ground potential, two ring-shaped electrodes at electrostatic potentials $V3$ and $V2$, and the mirror electrode with a potential of $V1$ as depicted in Fig. 2.3. The total electric field created by these electrodes acts as an electrostatic mirror and lens system for incoming electrons. The instrument is considered aberration-corrected when both, the sum of lowest rank chromatic aberrations of objective and mirror and the sum of third order spherical aberrations of objective and mirror are equal to zero, i.e. $C_{C}^{\text{total}} = C_{C}^{\text{obj}} + C_{C}^{\text{mirror}} / M^{2} = 0$ and $C_{3}^{\text{total}} = C_{3}^{\text{obj}} + C_{3}^{\text{mirror}} / M^{4} = 0$ where $M$ is the magnification in front of the mirror. The mirror has three variables - the electrostatic potentials $V1$, $V2$, and $V3$ - to adjust three optical parameters, namely $C_{C}^{\text{mirror}}$, $C_{3}^{\text{mirror}}$, and mirror focal length. All three parameters can be controlled independently. The scaling of the aberration coefficients with magnification is necessary since the aberrations of the objective lens depend strongly on image magnification $M$. The third order spherical aberrations scale with $C_{3} \propto M^{4}$ and the lowest rank chromatic aberrations scale according to $C_{C} \propto M^{2}$ [8]. Therefore, it is important that the image is placed in front of the mirror with a well-defined design-specific magnification [9]. The magnification can be adjusted by changing the sample-objective lens distance. Keeping the magnification close to the design value of $M = 8.5$ assures that the mirror can fully correct the aberrations within the limits of its finite working range (see Ref. [9] for a discussion on magnification measurements).

The optical performance of the electron mirror has been modeled by ray-tracing simulations [5]. These calculations show how the three mirror electrode potentials depend on each other (see Fig. 2.4a - voltages are indicated relative to a column potential of $-15010$ V) and they predict $C_{C}^{\text{mirror}}$ and $C_{3}^{\text{mirror}}$ as a function of $V1$, $V2$, and $V3$ (see Fig. 2.4b). The behavior of the mirror can be described approximately as follows: the voltage difference $V3 - V2$ is to first order proportional to the spherical aberrations of the mirror, $C_{3}^{\text{mirror}}$. The potential of the last electrode, $V1$, determines approximately the chromatic aberrations, $C_{C}^{\text{mirror}}$. The voltage $V2$ controls the focal length of the mirror. More precise control of the mirror aberrations is possible when they are considered to be functions of both $V1$ and $V3 - V2$ each, i.e. $C_{C}^{\text{mirror}}(V1, V3 - V2)$ and $C_{3}^{\text{mirror}}(V1, V3 - V2)$.

The question we want to answer here is if the two electron mirrors of ESCHER behave according to the theoretical predictions and if so how good is the agreement between theory and experiment? A quantitative answer is found by performing the following measurement: We pick a value for $V1$ and a value for $V3 - V2$ in
2.4. ABERRATION-CORRECTED LEEM IN PRACTICE

the range shown in Fig. 2.4a. The chosen voltage and voltage difference are applied to the respective electrodes of the mirror. We use now the potential \( V_2 \) to adjust focus of the mirror while \( V_3 \) follows since \( V_3 - V_2 \) is set to a fixed value. The in-focus condition of the mirror can be measured with the help of a TEM grid placed in the image plane in front of the mirror. Electrons coming from the sample form a shadow image of this grid before they go towards the mirror. Coming back from the mirror the same electrons form a second shadow image of that grid. The electron mirror is at in-focus condition if both shadows of the TEM grid are in-focus. This way we can measure \( V_2 \) as a function of sets of \( V_1 \) and \( V_3 - V_2 \) (see Fig. 2.4a). The measured sets of potentials can then be compared with the calculated values. Another way to look at Fig. 2.4a is that each data point corresponds to a different pair of \( C_{\text{mirror}}^c \) and \( C_{\text{mirror}}^s \) with the mirror at the in-focus condition. We find for the mirror on the right-hand side a perfect match with theory if \( V_1 \) is offset by \(-40\) V, i.e. a relative error of 0.24%. For the left-hand side mirror shifting \( V_1 \) by \(-27\) V and \( V_2 \) by \(+20\) V yields the theoretical behavior. The electron mirrors used in ESCHER, therefore, behave according to the ray-tracing results considering only small constant offsets as mentioned above. That means the calculated curves can be used in practice to control the first order properties of the electron mirrors. With the first order properties (Fig. 2.4a) accurately reproduced over a wide range of parameters, we can be confident that the chromatic and spherical aberrations (Fig. 2.4b) will also be accurately reproduced, as there is a 1 : 1 (but non-linear) relation between Fig. 2.4a and Fig. 2.4b. A set of electrode potentials to achieve a certain \( C_{\text{mirror}}^c \) and \( C_{\text{mirror}}^s \) can be obtained from these theoretical curves. Reference [9] discusses how the data in Fig. 2.4 is inverted to yield \( V_1, V_2, \) and \( V_3 \) as a function of \( C_{\text{mirror}}^c \) and \( C_{\text{mirror}}^s \).

2.4.2 MEASURING ABERRATIONS

For aberration-corrected LEEM and PEEM the mirror potentials have to be set such that both the total of chromatic aberrations \( C_{\text{C}} \) and the total of spherical aberrations \( C_{\text{3}} \) are zero. The aberrations are mainly due to the cathode objective lens and only minor contributions are caused by the other optical elements in the microscope. Therefore, aberrations are corrected to first order if the mirror is set up to introduce chromatic and spherical aberrations with same magnitude but opposite sign as the ones of the objective lens. The chromatic and spherical aberrations of the objective lens as a function of electron energy at the sample are well-known from ray-tracing calculations and from analytical theory [5, 10]. Then, a good first setting is obtained using the theoretical predictions of objective lens aberrations and mirror aberrations. To quantify the mirror setup we have to measure the chromatic and spherical aberrations of the microscope. Ways to measure chromatic and spherical aberrations of a LEEM have been introduced by Tromp et al. [8, 9].
In the following I will outline these measurement protocols and present aberration measurements performed with ESCHER.

**Chromatic aberrations**

Chromatic aberrations cause a displacement in the image according to

\[
\Delta C = C_C \left( \frac{\Delta E_0}{E} \right) \alpha + C_{CC} \left( \frac{\Delta E_0}{E} \right)^2 \alpha + C_{3C} \left( \frac{\Delta E_0}{E} \right)^3 \alpha^3 + \ldots \quad (2.1)
\]

with \(C_C\) the lowest rank chromatic aberration coefficient and \(C_{3C}\) and \(C_{CC}\) higher rank chromatic aberration coefficients.

In a non-corrected microscope the lowest rank chromatic aberrations shift the image plane according to \(\Delta C / \alpha = C_C \frac{\Delta E_0}{E}\), i.e. changing the electron energy will shift the image plane. Varying the lens current that excites the magnetic field of the objective lens changes focus and therefore also shifts the image plane. To determine the chromatic aberrations of the objective lens we can in principle measure the lens current necessary to offset the image plane shift caused by the chromatic aberrations as a function of electron energy. However, we need to be aware that the objective lens chromatic aberrations are the sum of aberrations caused by the strong electrostatic immersion field and by the focusing magnetic field. The lowest rank chromatic aberration, \(C_C\), can be treated as a superposition of the aberrations.
due to the electrostatic field $C_{Ce}$ and due to the magnetic field $C_{Cm}$ \[^{[8]}\]

\[
C_C = C_{Ce} + C_{Cm} = -L \sqrt{\frac{E}{E_0}} + C_{Cm} \tag{2.2}
\]

where $L$ is the distance between sample and anode of the cathode lens. $C_{Ce}$ depends on the electron energy at the sample $E_0$ and on the energy gained by electrons in the electrostatic immersion field $E$.

In an aberration-corrected microscope we have to add the chromatic aberrations of the aberration-correcting electron mirror $C_{Cmirror}$ to yield the total chromatic aberrations taking into account the scaling of the aberration coefficients with magnification

\[
C_C = C_{Ce} + C_{Cm} + \frac{C_{Cmirror}}{M^2} \tag{2.3}
\]

where $M$ is the magnification in front of the mirror.

The energy gained by the electrons in the electrostatic immersion field $E$ is determined by the bias voltage of the sample. The electron energy at the sample $E_0$ is determined by the difference between electron source potential $G$ and the sample bias voltage and is therefore given by $E_0 = G - E$. The total electron energy after acceleration is equal to the gun potential, $E_0 + E = G$.

If we vary the gun potential while keeping the sample bias constant we also vary the total energy after acceleration $E_0 + E$ and the electron energy at the sample $E_0 = G - E$. We are interested in the experimentally relevant case where $E$ is...
constant, and the electrons leave the sample with a start energy $E_0$, and an energy spread $\Delta E_0$. The image displacement due to such an energy offset $\Delta E_0$ can be measured by changing the gun voltage by $\Delta E$: $E_0 + \Delta E_0 = G + \Delta E - E$. Thus, the total chromatic aberration coefficient $C_C$ can be measured by varying the gun potential with fixed sample bias. It is, however, not very practical to perform such measurements because a change in gun potential also requires readjustment of the magnetic beam separators. For practical reasons it is therefore easier to measure $C_{Ce}$ and $C_{Cm} + C_{\text{mirror}}^m/M^2$ in two separate experiments. $C_{Cm} + C_{\text{mirror}}^m/M^2$ can be determined by an experiment that varies $E_0 + E$ and at the same time keeps $E_0$ at a constant value making the measurement insensitive to $C_{Ce}$. Such a scenario can be realized in a Hg PEEM experiment where $E_0$ is constant and determined by the photon energy and the workfunction of the sample. $E$ and therefore $E_0 + E$ can be varied by adjusting sample bias. The other part of the chromatic aberration coefficient, $C_{Ce}$, can be quantified by a typical LEEM experiment where the gun potential is fixed and the sample bias voltage is varied changing $E_0$ while $E_0 + E = G$ remains constant, i.e. the electrons pass through the magnetic part of the objective lens and through the mirror optics at fixed energy, and their chromatic aberrations are therefore not contributing to changes in lens current when $E_0$ is changed.

Figure 2.5a shows measurements of the objective lens current necessary to re-focus the image in Hg PEEM as we vary $E$. These measurements are repeated for different set values of the mirror chromatic aberrations $C_{\text{mirror}}^m$ (the required potentials are obtained from the data in Fig. 2.4). The slope $dI/dE$ changes sign as we change $C_{\text{mirror}}^m$ from 0 m to 10 m in steps of 2 m. These measurements were performed with $C_{\text{mirror}}^3 = -1900$ m. The authors of Ref. [9] show that the results in Fig. 2.5b are independent of $C_{\text{mirror}}^3$. The measurements are sensitive to the chromatic aberrations of the magnetic field of the objective lens and of the electron mirror. In Fig. 2.5b the slope is plotted as a function of the mirror chromatic aberration set value together with a fit to the data of the form $S = S_0 - aC_{\text{mirror}}$. In Fig. 2.6a measurements of the objective lens current in LEEM as a function of electron energy at the sample $E_0$ are plotted. The sum $E + E_0$ is kept constant making these experiments insensitive to the mirror aberrations and the chromatic aberrations of the objective lens’s magnetic field. The solid line in Fig. 2.6a is a fit of the form $I = I_0 + c\sqrt{E_0}$ [8]. The slope of this curve, i.e. the derivative $dI/dE_0 = \frac{c}{2\sqrt{E_0}} \propto C_{Ce}$, yields the electrostatic part of the chromatic aberration coefficient. The derivative is independent of the magnetic part of the chromatic aberrations of the objective lens and the chromatic aberrations of the electron mirror since the electron energy passing through these elements remain constant during these measurements.

The chromatic aberrations are corrected at one given electron energy $E_0$ if the
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Figure 2.6: (a) Objective lens current as a function of electron energy at the sample $E_0$ measured in LEEM. The solid line is a fit of the form $I = I_0 + cv/E_0$. (b) Chromatic aberration coefficient of the objective lens as a function of electron energy at the sample. The solid line obtained from measurements is in good agreement with theoretical values obtained using ray-tracing calculations (black circles).

sum of $C_{C}e + C_{Cm} + C_{C}^{\text{mirror}} / M^2 = 0$, i.e. if $c/(2\sqrt{E_0}) + S_0 - aC_{C}^{\text{mirror}} = 0$. This yields the required set value $C_{C}^{\text{mirror}}$ to correct the chromatic aberrations of the cathode objective lens at $E_0$. The chromatic aberration coefficient of the objective lens is then given by $C_{C}^{\text{obj}} = -C_{C}^{\text{mirror}} / M^2$, with $M$ the magnification in the object plane of the mirror. Figure 2.6b shows the measured chromatic aberration coefficient $C_{C}$ of the objective lens as a function of electron energy at the sample $E_0$ (solid blue line). Comparison with the theoretical values (black circles) obtained from ray-tracing yields a magnification of $M = 9$, close to the design value $M = 8.5$.

**Geometric aberrations**

If geometric aberrations are present, an electron ray leaving the sample at the optical axis under an angle $\alpha > 0$ will not be focused at the Gaussian image plane, but suffers a small displacement $\delta$. The displacement due to geometric aberrations is given by [8]

$$\Delta = C_1 \alpha + C_3 \alpha^3 + C_5 \alpha^5 + ... \quad (2.4)$$

with the objective lens defocus $C_1$, 3rd order spherical aberration coefficient $C_3$, and 5th order spherical aberration coefficient $C_5$.

Recently, Tromp et al. [9] suggested to measure the displacement due to spherical aberrations with the help of a sample with a rich diffraction pattern, like Si(111)-(7x7), in LEEM. We only illuminate a small spot with a size of about 200 nm diameter of the Si(111) sample. If the microscope were perfect and did not have any spherical aberrations we would see the 200 nm spot at in-focus condition as one
spot in the image. This image is, however, a superposition of all images coming from all the diffraction spots of the Si(111) sample at different $\alpha$. Now, if we defocus the objective lens of our perfect aberration-free microscope we would see not only one image of the spot but a multitude of images of this spot linearly displaced in radial direction with the displacement directly proportional to the defocus value (see Eq. 2.4). The objective lens of a real microscope, however, does have spherical aberrations causing an image displacement proportional to $\alpha^3$ (see Eq. 2.4) when $C_3$ is dominant. The total image displacement is then given by the sum of the contributions of third order spherical aberrations and defocus, neglecting the impact of higher order spherical aberrations. At in-focus condition the displacement of the small spot images is only caused by spherical aberrations. Over-focused small spot illumination images of Si(111)-(7x7) are shown in Fig. 2.7 for different set values of the mirror spherical aberrations $C_3^{\text{mirror}}$. The non-linear displacements decrease when going from $C_3^{\text{mirror}} = -500 \text{ m}$ (Fig. 2.7a) to $C_3^{\text{mirror}} = -1000 \text{ m}$ (Fig. 2.7b) and to $C_3^{\text{mirror}} = -2500 \text{ m}$ (Fig. 2.7c) compensating more and more for the spherical aberrations of the objective lens. The non-perfect rotational symmetry of these images is caused by residual tilt of the sample and electron beam as discussed in Ref. [11]. The data presented here were recorded with a previous version of a sample stage where sample tilt alignment was cumbersome and unreliable. With the recently installed improved sample stage, tilt alignment can be achieved in a much more precise and reliable fashion. A quantitative measure of the spherical aberrations can be obtained from these images by plotting the image displacement as a function of the known diffraction angles $\alpha$ of the corresponding diffraction spots. The displaced images are labeled as indicated in Fig. 2.7d. From ray-tracing we know that $C_3$ of the objective lens is about 0.5 m at a magnification of $M = 1$ at $E_0 = 1 \text{ eV}$ (see chapter 3). The largest diffraction angle considered in the aberration coefficient measurements here is about 20 mrad. The magnitude of the image displacements due to $C_3$ is therefore to be expected in the order of a few $\mu\text{m}$.

Figure 2.8a shows data obtained from an image taken with $C_3^{\text{mirror}} = -1000 \text{ m}$ at an electron energy of $E_0 = 5 \text{ eV}$. A fit of the form $\delta = C_1 \alpha + C_3 \alpha^3$ to the data gives the defocus $C_1$ and the total spherical aberration coefficient $C_3$, i.e. the sum of spherical aberration coefficients of the objective lens and the electron mirror. Figure 2.8b shows the measured $C_3$ as a function of mirror set value $C_3^{\text{mirror}}$. The error bars indicate the 95 % confidence intervals of $C_3$ obtained by fitting the displacement vs. diffraction angle data as shown in Fig. 2.8a for $C_3^{\text{mirror}} = -1000 \text{ m}$. The deviation of the data from the linear fit in Fig. 2.8b is mainly due to residual sample and beam tilt that give rise to asymmetric image distortions.

In Fig. 2.8b $C_3$ is referenced to an image plane with magnification $M = 1$. The nominal magnification of the object plane in front of the mirror is $M = 8.5$. The
ratio $\frac{C_{3\text{mirror}}}{C_3} = M^4$ is therefore expected to be 5220. The $C_3$ aberrations of the objective lens are corrected if $C_{3\text{mirror}} = -C_3 M^4$. The inverse of the slope of the linear fit in Fig. 2.8b is $5880 = M^4$. From these measurements we conclude that the magnification of the mirror object plane in our experiment is $M = 8.8 \pm 0.5$ - very close to the nominal design value. Furthermore, we find that the spherical aberrations are corrected at $C_{3\text{mirror}} = -3055 \text{ m}$ for an electron energy at the sample of $E_0 = 5 \text{ eV}$. Since the spherical aberrations of the objective lens depend on $E_0$ (see chapter 1) the measurement of $C_3$ has to be repeated at different electron energies (see Ref. [9]).

The measurements presented above show that chromatic and spherical aberrations can be corrected and quantified in ESCHER. What's more, using the outlined procedures to control spherical and chromatic aberrations we have been able to achieve a record spatial resolution of 1.4 nm of a SiC(0001) sample with graphene (see below and Fig. 2.9).

2.5 **Resolution**

So far, the best resolution obtained in an aberration-corrected LEEM experiment is approximately 2 nm [5]. Experiments with graphene grown on SiC(0001) show that it is possible to image with ESCHER with a lateral resolution $< 2 \text{ nm}$ during routine operation (see Fig. 2.9). The best lateral resolution obtained with ESCHER is 1.4 nm measured on a graphene covered SiC(0001) sample (see Fig. 2.9). The resolution is obtained by applying a 84 % - 16 % intensity criterion (see chapter 3). These high resolution measurements are experimental proof that chromatic and spherical aberrations are successfully corrected in ESCHER. Furthermore, it corroborates the validity of the measurement protocols to determine and control $C_C$ and $C_3$ in LEEM as discussed above.

In order to reach even higher resolution, several measures have to be taken to reduce the level of vibrations introduced into the system and to increase mechanical stability. First, we have developed a new sample stage. This new stage is directly attached to the objective lens, eliminating any direct mechanical coupling to other parts of the vacuum system. In addition, we have incorporated a mu-metal shield around the sample to suppress electromagnetic disturbances. In the vicinity of the sample, shielding is most important since the electron energy is at its lowest value. In addition, the vibration and the acoustic isolation of the microscope have to be optimized. The system is already installed on its own vibration-isolated building foundation and is equipped with an active vibration isolation system (AVI-400/LP and AVI-350/LP from TABLE STABLE, Switzerland). Vibrations transmitted through incoming wires, tubes, and vacuum pumps are
Figure 2.7: Over-focused LEEM images of Si(111) taken with a small illumination spot for different set values of the spherical aberrations of the aberration-corrector. The non-linear displacement decreases with increasingly corrected $C_3$ (see dashed lines). Distortions due to residual sample and beam tilt make the images non-rotational symmetric.
2.5. Resolution

minimized by passive vibration isolation. Moreover, acoustic isolation becomes increasingly important to reach the ultimate resolution. Other sources of interference include alternating-current magnetic fields that are present in the vicinity of the microscope. Even small fields can cause oscillatory displacements of the image and therefore degrade the resolution. This can be overcome by improving the passive magnetic shielding, particularly of the sensitive magnetic prism arrays. Furthermore, the stability of the power supplies of the microscope needs to be improved by about an order of magnitude to better than 0.1 ppm. Finally, the image detection system has to be improved to reach a resolution as low as 1 nm at a field of view of 1 µm, i.e., the detector must be able to resolve 1000 pixel elements across the field of view. MCP/phosphor/charge-coupled device camera detector systems currently in use in most LEEM/PEEM instruments have a spatial resolution of about 130 µm across a 40-mm diameter detector. This gives only 300 resolvable pixel elements across the detector. An attractive replacement of this commonly used detection system is the Medipix detector - a solid-state pixel detector. In the framework of the ESCHER project, we have shown that the detector resolution of Medipix is better by about a factor of 2, as compared with a standard MCP-based detector [12]. Figure 2.10 shows LEEM images of graphene flakes grown on an Ir(111) substrate recorded with the LEEM setup at the University of Twente. The image in Fig. 2.10a was recorded with a conventional MCP detection system, and the image in Fig. 2.10b was recorded with a Medipix2 detector. Both images show the same sample area at identical electron-optical magnification. It is clear from these images that finer image details are visible in the image taken.
Figure 2.9: LEEM images of graphene grown in-situ on SiC(0001) samples. Field of view is 660 nm. (a) A line scan along the red rectangle with a width of 30 profiles is shown at the right. Applying a 84% - 16% intensity criterion to the step in this profile gives a spatial resolution of 1.4 nm. (b) The two line profiles (average over 5 profiles) on the right yield a resolution of 2 nm and 1.6 nm.
2.6 Summary

We have described the layout and the capabilities of a new aberration-corrected LEEM and spectroscopy facility, i.e., ESCHER, at Leiden University. Initial experimental results obtained on thin graphene films grown in situ on SiC(0001) at 1700 K are very promising and demonstrate that the electron optical system is fully operational. In our quest for the ultimate spatial resolution, we have demonstrated successful correction of chromatic and spherical aberrations yielding a record resolution of 1.4 nm measured on graphene grown on a SiC(0001) substrate. We have addressed the various factors that limit the experimental resolution and have discussed what measures we have taken and what advancements are still needed to improve it toward the theoretical limit of about 0.5 nm. In the following chapter 3 we will introduce a theory that allows one to calculate the limit resolution theoretically achievable with our aberration-corrected LEEM instrument.

Figure 2.10: LEEM images of graphene flakes on Ir(111) recorded with (a) a conventional MCP detector and (b) a Medipix2 detector with the LEEM setup at the University of Twente. Both images show the same sample area at identical magnification with a field of view of 1.5 µm. The detector resolution is improved by about a factor of 2 in the case of Medipix2.
REFERENCES


