Low-energy electron microscopy on two-dimensional systems : growth, potentiometry and band structure mapping

Kautz, J.

Low-Energy Electron Microscopy on Two-Dimensional Systems:

Growth, Potentiometry and Band Structure Mapping

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The cover shows an artist's impression of the potential map in Fig. 5.4.

Preface

Most textbooks on Low-Energy Electron Microscopy (LEEM), will start by describing it as the ideal tool for surface science. And while it is true that the short mean free path of the low-energy electrons makes the technique very suitable to study surface structures, the focus of the LEEM community has significantly broadened over the years. What started out as a technique with a narrow field of application quickly grew into a versatile many-purpose research tool. It is one of the goals of this work to extend the applicability of LEEM in condensed matter physics even further.

The insight that LEEM can be used for more than surface science is not new. Already since the creation of the first LEEM by Telieps an Bauer [1] it was clear that there was a relation between the electron reflectivity and electronic states in the studied system. Still, for a long time there has been a focus on the analysis of surface structures. This is mainly because of the ample opportunities available for LEEM in this field of research. The invention of LEEM suddenly gave direct access to the dynamics of surface processes, which up till then only could be inferred. Many fundamental surface science problems, like the dynamics of surface reconstructions and layer growth, could suddenly be solved due the direct visualization by LEEM [2–4]. However, these early successes sparked an increased interest in LEEM and as the LEEM community grew, its scope would broaden.

Parallel to the physics research, there has been a lot of work on the improvement of the spatial resolution of LEEM instruments. Where the first instruments had a resolution of $\approx 20\,\mathrm{nm}$ [1], the present day optimal resolution, as obtained in the Leiden ESCHER set-up described in chapter 1, is 1.4 nm. One key ingredient that enabled the enormous improvement in resolution is the implementation of aberration correction. Aberration correction, which was already applied in Transmission Electron Microscopy (TEM) by the use of a series of monopoles [5], was introduced for LEEM on the basis of an electrostatic mirror [6–8]. Aberration correction alone has improved the resolution of LEEM by a factor 5 [9].

Technical developments have also played a great role in the diverging of LEEM applications from pure surface science. The broadening of the scope of LEEM went hand in hand with the development of new imaging methods. One big step in this evolution was the implementation of energy filters in LEEM instruments. This made it possible to combine LEEM, Low-Energy Electron Diffraction (LEED), Angle Resolved Photo-Emission Electron Microscopy (ARPES) and Electron Energy Loss Spectroscopy (EELS) measurements in one instrument, a unprecedented combination of powerful surface analysis tools.

The development of new LEEM-based techniques is still ongoing. The realization of spin-polarized electron sources with an ever increasing polarization[10] has opened the road for the study of magnetic domains[11,12]. Moreover, the development of so-called pump-probe experiments allows analysis of dynamics on femtosecond timescales[13,14]. In this work we add yet two more techniques to this already broad repertoire of analysis tools.

This thesis starts with an introduction of LEEM in general, the various available imaging modes and a description of our aberration-corrected ESCHER set-up in chapter 1. The rest of this thesis consists of two parts which follow a similar pattern as the history of LEEM. In the first part (chapters 2 and 3) we will use LEEM in the traditional way to study growth and analyze surface structures. Chapters 4 to 6 form the second part, where we move away from pure surface science and introduce two new LEEM techniques.

We start by investigating (sub)monolayers of Au on Si(111) in chapter 2. Specifically, we study the (5x2) structure, which is formed at low Au coverages and exhibits metallicity in only one direction. There has been a lot of controversy over the exact atomic configuration of this surface structure[15–17]. Our results show that the number of of Au atoms per unit cell is different than previously assumed and that a revision of the structural model is necessary. In chapter 3, we analyze how alkanethiols react with the (sub)monolayers of Au on Si(111), studied in chapter 2. The growth of alkanethiols on bulk Au crystals has been studied extensively[18–20]. The stability of the self-assembled monolayers that form is mainly due to the strength of the S-Au bond[19]. Based on the results obtained on bulk crystals one would naively expect that when submonolayers of Au are exposed to alkanethiols, the alkanethiols will react with those areas of the surface with the highest Au concentration. Surprisingly, our results show something completely different.

In the second part of this thesis, we introduce two new techniques. First, in chapter 4, we demonstrate a completely new method to analyze electron transport using LEEM. Electron transport measurements are typically performed in a two- or four-probe configuration. This allows one to perform average conductivity measurements, but local variations in conductivity are lost. By employing low-energy electrons to locally probe the electric potential, while applying an in-plane bias voltage, we are able to create potential maps of the surface. This enables us to locally probe the conduction properties of a sample. We demonstrate our new technique, coined Low-Energy Electron Potentiometry (LEEP), by visualizing Schottky behavior at the Au-Si interface. In chapter 5, we show that for layered materials, the sensitivity of LEEP can be drastically improved. The coupling of the imaging electrons to interlayer states causes pronounced features in the energy dependence of the reflectivity [21,22]. We can use these features to improve the accuracy of the local potential measurements. Finally, in the last chapter, we study the influence of non-normal incidence on our potentiometry results. Interestingly, we find that we can use the in-plane-momentum dependence of the electron reflectivity to obtain information on the interlayer states. This allowed us to map out the band structure of these unoccupied interlayer states in graphene. We expect this technique to be particularly useful for the analysis of van der Waals materials [23]. With these new techniques, in combination with the already wide range of existing analysis tools, LEEM provides an unparalleled experimental set-up for complete in-situ sample characterization.

Jaap Kautz
Leiden, March 2015

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