Surface Electron Microscopy of Ga Droplet Dynamics on GaAs (001)

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We describe the design and application of a surface electron microscope which has been constructed to study the *in situ* growth dynamics occurring on III-V semiconductor surfaces during molecular beam epitaxy. Mirror electron microscopy and photo-emission electron microscopy (PEEM) are used to elucidate the physics of Langmuir (free) evaporation of GaAs. Of particular interest is the formation and motion of Ga droplets across the surface. We also describe the surface electron microscopy technique known as Lloyd’s Mirror PEEM which has been developed to image surface topography in real-time. This is applied to determine Ga droplet contact angles during evaporation and the extension to synchrotron based Lloyd’s Mirror x-ray PEEM (XPEEM) is considered.

**Keywords** photo-emission electron microscopy; mirror electron microscopy; gallium arsenide; gallium droplets; Langmuir evaporation; Lloyd’s Mirror

1. Introduction

GaAs based communication devices are now ubiquitous in everyday life. Applications range from mobile phones and satellite communications to wireless networks and automotive collision warning systems. The direct-band gap property of GaAs has also opened up numerous applications in optoelectronics including laser pointers, printers, barcode readers and DVD players. The advances in radio frequency (RF) communication technology have chiefly arisen from the development and availability of microwave monolithic integrated circuits or MMIC’s. The MMIC has had the same impact in the expansion of modern communications technologies as silicon integrated circuits made in the advancement of the computer industry. Just as the Si transistor is the main building block of the integrated circuit, MMIC’s are composed of thousands of GaAs high electron mobility transistors (HEMTs). GaAs transistors therefore play a foundational role in modern communications. Similarly, the III-V based laser diode can be viewed as the major component underlying the optoelectronics industry. A key similarity between HEMTs and laser diodes is that they are composed of thin layers of III-V semiconductor material, the thicknesses of which must be finely controlled. This is achieved by growing such layers via molecular beam epitaxy (MBE) with atomic layer precision. However, despite the technological importance of controlling III-V MBE growth, the real-space imaging of surface growth dynamics has remained elusive. This is primarily because the incident As flux plays a key role in influencing the surface thermodynamics [1] and any technique must be able to cope with this flux as well as image the substrate surface at high temperature under ultra high vacuum (UHV) conditions.

In addition to improving and advancing current technologies, the long-term research objective in III-V materials is to fabricate new quantum structures of nanoscale dimensions. Utilising variants of MBE, it has been possible to move beyond the limitations imposed by conventional lithography and assemble a variety of structures including quantum dots, double dots, rings, double rings, molecules and rods [2-4]. Potential applications include novel lasers, optical communications within chips, electron spin memory and quantum computing and cryptography. Such tiny quantum structures also offer new opportunities for studying fundamental physics such as the Aharonov-Bohm effect. However, again a key limitation in the control and development of quantum structures is our inability to see how they form in real-time. As in the case of interfaces grown by MBE, quantum structure formation is a dynamic process occurring in UHV under As flux at elevated temperatures. Being able to understand how atoms collectively rearrange on the nanoscale under real growth conditions would facilitate the understanding of the basic science underlying quantum structure formation and provide new insight into how to fabricate new structures.

To address the technologically important issues of imaging interface and quantum structure formation under As flux we have developed a novel instrument which combines a surface electron microscope with a III-V MBE growth system. In this chapter we will discuss the basic design and capabilities of this III-V low energy electron microscope (LEEM). Applications of III-V LEEM to the study of (free) evaporation of GaAs into a vacuum are then reviewed, revealing the unexpected and striking motion of Ga droplets. We will also describe the recently developed technique of Lloyd’s Mirror photo-emission electron microscopy (PEEM) which can be used to map surface topography and quantify the morphological evolution of surfaces [5, 6].
2. III-V LEEM

2.1 Electron optics and imaging modes

The electron optics design is based on the standard Elmitec LEEM III configuration. However, the system incorporates a Schottky field emission electron source for enhanced brightness and beam coherence compared with the conventional LaB$_6$ gun. A schematic of the electron optics is shown in Fig.1. Electrons with energy ~20 kV are transferred from the Schottky source to the back-focal plane of the objective lens via a series of condenser lenses (CL1-CL3) and a magnetic sector which deflects the beam. The objective lens is the heart of the LEEM system and combines a magnetic focusing field inside the lens with an electrostatic field between the lens and sample. An enlarged view of this region is shown in inset contained in Fig. 1. The sample is maintained at a potential close to that of the electron source with the objective lens at ground potential. The incident beam therefore traverses the magnetic part of the lens at relatively high voltage (~20 kV) but is decelerated in the 2 mm gap between objective lens and sample to an energy of 0-100 eV. Following the low energy interaction and reflection from the sample surface, the electrons are re-accelerated to the gun energy on their return to the objective lens. The magnetic sector then deflects the electrons into the projector column where an image is formed using a channel plate and CCD camera.

The flexibility of the optical system is a major advantage which provides several imaging possibilities [7-9]. In LEEM, a parallel, coherent beam of low energy electrons (0-100 eV) is focussed onto the specimen and a 4 nm resolution image of the surface is formed from the elastically back-scattered electrons. This is sufficient to study the nanoscale self-organisation processes of interest during semiconductor growth. LEEM has superb surface sensitivity due to the very low energy of the incident electrons and can acquire images over a large field of view at video rates. It is therefore well suited for the real-time, non-invasive study of surfaces and interface formation during MBE growth. The back-scattered electrons form a diffraction pattern in the back-focal plane of the objective lens and an aperture can be placed around appropriate spots to obtain contrast from reconstructed surface domains. Alternatively, surface steps can be imaged directly using phase contrast [10-12]. In the mirror electron microscope (MEM) mode the sample potential is slightly more negative than the electron source potential [13-15]. In this case, the electrons are reflected in front of the specimen and do not make contact with the surface. Image contrast results from small perturbations in the otherwise homogeneous applied electric field between the objective lens anode and the sample cathode. This can result from surface topography or electric field variations due to work function differences or potential differences in electronic components. Additionally, in the PEEM mode, electrons are excited by incident photons and are emitted from the surface and imaged in the projector column [16].

Fig. 1 Schematic of the electron optics of the III-V LEEM. The illumination column consists of three condenser lenses (CL1-3). The electron beam is deflected by a magnetic prism and focussed by the objective lens onto the sample. An enlarged view of the objective lens region is shown in the inset. Reflected electrons are again deflected by the prism into the imaging column which consists of transfer (TL), field (FL), intermediate (IL) and two projector (P1, P2) lenses.
2.2 Modifications for III-V MBE

The incorporation of III-V MBE required significant modification to the basic LEEM system including the installation of multiple deposition sources, dedicated equipment for surface cleaning and an internal cooling shroud to limit the build-up of As background pressure. Figure 2 contains an overview of our instrument with corresponding cross-sections of the basic LEEM system and the specimen region shown in Figs. 3(a) and (b), respectively.

Our III-V MBE sources (MBE-Komponenten) consist of Ga and In effusion cells with integrated cooling shrouds and shutters and a metal valve As cracker source. The effusion cells are mounted in a face-up configuration (underneath the instrument in Fig. 2) since Ga and In are liquids at high temperature. Crucibles of 10 cc capacity are heated by dual filaments in a hot-lipped configuration to minimise condensation and droplet formation near the orifice. This improves sample quality and flux reproducibility. Both sources are equipped with automatic shutters. A PBN cap with small orifice together with additional Ta shielding ensures a well-collimated evaporation beam to reduce material deposition in the UHV chamber and the objective lens region in particular. The Ga source is equipped with a UPS backup system to maintain the source temperature at 50 °C in the event of a power failure. This prevents the cracking of the crucible due to Ga expansion on freezing.

The As cracker source consists of a 300 cc As reservoir built into a water-cooled vacuum enclosure. The reservoir is heated to evaporate the As and a cracking zone on top of the reservoir facilitates the conversion of naturally sublimated As$_4$ into As$_2$ so that the deposition species can be controlled. An integrated all metal valve separates the reservoir and cracking zone providing precise As flux control. The reservoir and cracker temperature gradient is computer controlled during heating and cooling to reduce stress on the valve. Water flow meters are installed to control and optimize flow to all sources. The source temperatures and the background vacuum pressure are continuously monitored.

The III-V LEEM system incorporates a cooling shroud to reduce the background As pressure resulting from high molecular As$_4$ or As$_2$ flux from the cracker source. This is critical in surface electron microscopy to prevent discharge between the specimen and objective lens. A detailed drawing of the shroud design is contained in Fig. 3(b). The Cu shroud is mounted inside the UHV chamber and liquid nitrogen (LN) is supplied via vacuum feedthroughs to achieve thermal insulation. Small apertures in the shroud allow Ga, In and As flux to impinge on the centre of the sample. Typically, the background pressure is reduced by nearly four orders of magnitude from 5x10$^{-5}$ to 2x10$^{-9}$ T when the shroud is in operation which provides suitable conditions for imaging. After long time exposure to As flux, the objective lens surface eventually becomes rough due to material deposition. A regular cleaning procedure is therefore necessary to reduce the probability of discharge. Magnetic shielding is installed on the sample manipulator flange.
surrounding the cathode and anode region in Fig. 3(b) to screen the electron beam from stray fields originating from the MBE sources.

![Fig. 3](image)

(a) Cross-section of the LEEM instrument. (b) An enlarged view of the objective lens area showing the location of the cooling shroud and access for the MBE sources.

To obtain clean and atomically flat GaAs surfaces is essential for LEEM studies. As an additional tool for in situ surface cleaning, we employ an atomic hydrogen source (AHS) [17]. A beam of atomic hydrogen is created by cracking hydrogen gas molecules (H$_2$) in a tungsten tube heated by electron bombardment up to 2200 °C. The AHS is effective at removing carbon and other surface impurities as well as oxides. Atomic hydrogen reacts with Ga$_2$O$_3$ of the epikready GaAs surface to create the more volatile compound Ga$_2$O via, Ga$_2$O$_3$ + 4H = Ga$_2$O ↑ + 2H$_2$O↑. The Ga$_2$O desorption then leaves the surface devoid of the oxide without damaging surface morphology.

3. Langmuir evaporation of GaAs (001)

3.1 Imaging Ga droplets above the congruent evaporation temperature $T_c$

Langmuir (free) evaporation of GaAs (001) into a vacuum is a classic topic which has been studied for many decades [18-21]. Although this is perhaps the simplest experiment one can perform in the III-V LEEM, a real-space imaging capability still provides a number of unexpected surprises. When GaAs is heated in a vacuum it decomposes and As and Ga fluxes evaporate from the surface (Fig.4). It is known that below a certain temperature $T_c$, known as the congruent evaporation temperature, the fluxes are equal and compound stochiometry is preserved. However, above $T_c$, As evaporates more readily leaving behind Ga rich droplets on the surface [22, 23]. Presently, there is significant interest in such droplets because by recrystallising them under As flux one can generate new quantum structures in a process termed droplet epitaxy [2, 24-26]. Such structures can now be formed in complex configurations including multiple-ring structures [24, 26] and, as discussed in the introduction, have many and varied potential applications.
Fig. 4 Left: Ratio between As ($F_{\text{As}}$) and Ga ($F_{\text{Ga}}$) fluxes leaving GaAs (001) during Langmuir evaporation. The open circles were obtained by mass spectrometry measurements [18]. Below the congruent evaporation temperature $T_c$, $F_{\text{As}} = F_{\text{Ga}}$ and above $T_c$, $F_{\text{As}} > F_{\text{Ga}}$ leaving behind Ga-rich liquid droplets.

To study the decomposition of GaAs we can therefore heat an epi-ready GaAs substrate above $T_c$ and image droplet formation in the III-V LEEM. MEM is an ideal imaging mode for studying surface morphology and an image of Ga droplets is contained in Fig. 5. The droplets appear as dark circles enclosed by a bright-concentric ring. This contrast can be understood from a numerical evaluation of classical electron ray trajectories. Fig. 6(a), for example, shows the ray density in the MEM image plane as a function of objective lens defocus. The droplet distorts the uniform electric field between the planar sample surface and the objective anode which significantly redistributes electron intensity. A horizontal cut across the ray trajectories maps the image intensity for a given defocus. The envelopes of rays contained in Fig. 6(a) can be viewed as caustics and the MEM image is therefore a projection of these caustics on the image plane.

For positive defocus, image intensity is dominated by a strong central caustic (Fig. 6(a)) surrounded by a dark region denuded of rays. However, this bright central contrast fades close to $\Delta f=0$, and for negative defocus it is replaced by a central dark region surrounded by a bright caustic ring which is a convenient condition for imaging droplets. The distance between adjacent rays can be converted into an image intensity as shown in Fig. 6(b) for a defocus $\Delta f = -20 \mu\text{m}$, which is similar to the experimental contrast observed in Fig. 5. It should be noted that the projected caustic ring diameter in Fig. 6(a) is larger than the actual perimeter of the droplet so that the droplet size is enlarged in MEM.

Fig. 5 MEM image of Ga-rich liquid droplets on GaAs (001) at 630 °C (scale bar, 2 µm). The droplets appear as dark circles surrounded by bright concentric rings and are somewhat larger than the actual droplet size (see Fig. 6(a)).
Fig. 6 (a) Ray densities in a cross-section of the objective lens image plane as a function of defocus $\Delta f$. The perimeter of the droplet is indicated by the vertical dashed lines. The 1D cross-sectional image intensity for a given $\Delta f$ is evaluated from the distance between adjacent rays on a horizontal line, parallel to the $x$-axis. (b) Image intensity for a defocus $\Delta f = 20 \mu m$ which is similar to the experimental contrast displayed in Fig. 5.

3.2 Running droplets of Ga during evaporation

The most striking observation in the III-V LEEM is that Ga droplets run across the surface spontaneously during annealing. Motion continues, even after the evaporation of many hundreds of atomic layers of the crystal. The formation of droplets during Langmuir evaporation has been studied for many decades but, to our knowledge, droplet dynamics has not been observed previously, illustrating the advantages of in situ real-time imaging.

![Fig. 7](image)

**Fig. 7** MEM images captured from a movie of droplet surface dynamics [27]. The reference arrow marks the position of two stationary droplets at the start of the sequence. The droplets grow in size with time and move along [110] directions.

An example of droplet motion is illustrated in Fig. 7. The epitaxy-ready surface is associated with a slight roughness, and as the droplets move they leave behind a smooth trail. The motion has a stick-slip character and occurs preferentially along the [110] direction, but equally in both directions, ruling out thermal gradient effects. The motion can be explained by considering the net force on the droplet derived from interfacial energies which in turn depend on local chemical potentials. Considering the thermodynamics of the GaAs surface during evaporation one can obtain an expression for the total net force on a droplet of diameter $d$ at temperature $T$ as

$$ F = \alpha (T - T_*)^2 d, $$

where the coefficient $\alpha$ embodies both thermodynamic and kinetic properties of the surface [27]. This is a surprising prediction as motion is expected at temperatures well above and well below $T_*$ but not at $T_*$ itself.

To test this relationship we heated a sample above $T_*$ to form droplets and then adjusted the temperature to the desired $T$ and measured the droplet velocity averaged over many minutes. The results are contained in Fig. 8. It can be seen that there is a temperature range of approximately 20 °C about $T_*$ where the droplets do not move. However, away from $T_*$, the average droplet velocity increases with either increasing or decreasing temperature. In addition,
moving droplets at high $T$ grow with time whereas moving droplets at low $T$ shrink. This is expected from droplet stability arguments and independently confirms that the temperature where motion ceases is set by $T_c$.

The analysis can be taken further by modelling the time-averaged stick-slip motion as a damped response to $F$ with an effective frictional force which is independent of velocity and opposite to the direction of motion. From Eq. (1), the velocity then becomes

$$v \approx m\alpha(T - T_c)^2 - \nu_f,$$

where $m$ is the mobility and $\nu_f$ the friction term. When $(T - T_c)^2 < \nu_f / ma$ then $v = 0$ which explains the region of zero motion about $T_c$. When the range of $T$ is large it is necessary to include cubic and higher order terms in the expansion about $T_c$ and with Eq. (2) extended to cubic order it is possible to fit the velocity data well, as shown in Fig. 8. The basic prediction of $v \propto (T - T_c)^2$ is, however, sufficient to capture the overall general behaviour.

![Fig. 8](image_url) Average droplet velocity plotted against temperature $T$ [27]. The solid line is the truncated cubic fit and the dashed line is the quadratic term alone which has been extracted from the same fit. The error bars represent the scatter of individual measurements made at a given $T$.

The mechanism for running droplets described in this section requires only that, at temperatures where a liquidus exists, the more slowly evaporating component forms droplets on the surface. Therefore, one might anticipate similar behaviour in other III-V semiconductors such as InAs and possibly in many other systems. Furthermore, the intrinsic droplet motion may open up new extensions for the droplet epitaxy technique.

4. Topographic mapping using surface electron microscopy

4.1 Mapping of surface topography

In the study of Langmuir decomposition it would be advantageous to determine Ga droplet morphology. It is known that Ga droplets etch the GaAs substrate during evaporation [23] and the contact angle geometry is important for understanding the forces acting on the droplet. However, a major challenge in surface electron microscopy is the ability to extract 3D topographical data. We have therefore developed the Lloyd’s Mirror PEEM technique [5, 6] as a means of extracting 3D information from plan-view images. The method conforms to the standard geometry for threshold emission PEEM and so it is directly complementary to other surface electron microscopy imaging modes such as LEEM, MEM and conventional PEEM.
4.2 Lloyd’s Mirror PEEM

The classic optics experiment known as ‘Lloyd’s Mirror’ was developed by Humphrey Lloyd in the 19th century. The basic principle is illustrated in Fig. 9. A light source A is used to form an interference pattern on a distant screen by superposing direct illumination from the source with its reflection in a plane mirror at O [28]. This simple geometry has seen a range of important applications including astronomical interferometry [29] and high resolution lithography [30]. We can also recreate Lloyd’s Mirror in surface electron microscopy by replacing the light source with a mercury lamp, the mirror with a planar substrate and the screen with a 3D surface object of interest, such as a Ga droplet. Lloyd’s fringes projected onto the object will therefore modulate electron photoemission which can be imaged in the III-V LEEM [5, 6].

The detailed set-up for Lloyd’s Mirror PEEM is illustrated in Fig. 10. A surface object (e.g. a Ga droplet) is illuminated by a beam of incident radiation which is defined by an aperture A of radius R. For simplicity, first consider a single monochromatic point source located at S within the aperture plane. This source illuminates the object at position F both directly and indirectly from its reflection in the substrate surface at P, forming fringes on the object surface. This is similar to Young’s double slit interference experiment arising from interference between the source at S and its virtual image at S’. However, the observed intensity is the inverse of the expected Young’s fringes because a phase shift of π is induced by reflection from the substrate.

In practice, the idealized monochromatic source S is polychromatic and we must also take into account the spatial incoherence introduced by the finite aperture A. The intensity at the specimen surface is then given by

\[ I_p\left(\mathbf{k}, \Delta k, R, L, z\right) = \frac{4\Delta L \phi^2 I_s^p\left(\mathbf{k}, \Delta k, \phi, z\right)}{\pi R^2 \sqrt{R^2 - L^2 \phi^2}} d\phi, \]

where L is the distance between the aperture and the object. The function \( I_s^p\left(\mathbf{k}, \Delta k, \phi, z\right) \) is the polychromatic intensity from a rectangular slit of small angular size \(2\phi\) (see Fig. 10):
In the practically significant case of threshold photoemission, the electron excitation is dominated by a single broadened emission line of wavevector \( \vec{k} = \frac{2\pi}{\lambda} \) at mean wavelength \( \lambda \) and Gaussian shape of FWHM \( \Delta k \approx \frac{\Delta \lambda}{\lambda} \), where \( \alpha \) is the angle of incidence of the radiation on the surface within an assumed paraxial approximation. Numerical evaluation of Eq. (3) indicates that, provided the illumination aperture diameter is appreciably smaller than the aperture–object distance, the fringe intensity at a specific position on the object surface depends only on the \( z \)-coordinate (height) of that position. In particular, the height difference \( \Delta z \) between fringe intensity maxima is, to a good approximation, given by

\[
\Delta z = \frac{\lambda}{2 \sin \alpha}.
\]  

Eq. (5) indicates that Lloyd’s fringes can be used as a basis for determining the shape of a surface object.

We must now relate the UV interference pattern at the object surface to the Lloyd’s fringe pattern observed in the electron microscope. The mean free path of electrons in the solid \( \ell \) is restricted to several nanometres [31] for incident UV radiation energy, which is only slightly greater than the work function. As the Lloyd’s fringe spacing \( \gg \ell \), we may therefore assume that all of the emitted electron flux is proportional to the incident radiation \( I_p(z) \), given by Eq. (3). However, it is well-recognized that non-uniform surface topography deforms the uniform electric field between the anode and sample which influences the photo-emitted electron trajectories [32, 33]. The resulting shifts in Lloyd’s fringes must therefore be taken into account when reconstructing surface topology via Eq. (5).

To evaluate the fringe shifts we use the approach of Nepijko et al. [32, 33] who determine the shift (in surface coordinates) to be:

\[
S_x(x, y) = \frac{1}{E_0} \int_0^\infty \frac{\partial \Phi(x, y, z)}{\partial x} d\tilde{z},
\]

where \( \Phi(x, y, z) \) is the potential distribution above the object surface and \( E_0 \) is the uniform electric field that accelerates the electrons across the specimen-anode aperture gap. A similar expression exists for \( S_y(x, y) \). Surface roughness \( h(x,y) \) produces an electric field perturbation in its vicinity, which is equivalent to the field from a planar potential distribution \( \Phi(x, y, z = 0) = E_0 h(x, y) \). The potential is then determined from the solution to Laplace’s equation, \( \nabla^2 \Phi(x, y, z) = 0 \), subject to this planar boundary condition. This is the corresponding Dirichlet problem for a half-space, \( z > 0 \), whose solution is given by:

\[
S_x(x, y) = \frac{1}{2\pi} \int_{\infty}^{\infty} \frac{\tilde{h}(x - \xi, y - \eta)}{\left(\xi^2 + \eta^2\right)^{3/2}} d\tilde{\xi} d\tilde{\eta},
\]  

where the integration variables \( \xi \) and \( \eta \) correspond to the \( x \) and \( y \) coordinates, respectively. If the surface potential depends only on the \( x \) coordinate it can be shown that the electron shift \( S_x \) is the Hilbert transform of the object height function \( h(x) \) [33]:

\[
S_x(x) = H[h(x)] = -\frac{1}{\pi} \int_{-\infty}^{\infty} \frac{h(\xi)}{x - \xi} d\xi.
\]

For the general and 1D cases respectively, Eqs. (7) and (8) provide a means of correcting for the fringe shifts and reconstructing the surface shape via Eq. (5).
4.3 Analysis of the rectangular Ga droplet morphology

Upon annealing the GaAs (001) surface above the congruent evaporation temperature to \( \sim 660 \) °C, a rectangular-based droplet morphology was sometimes observed by in situ PEEM (Fig. 11(a)) and ex situ atomic force microscopy (AFM) following quenching (Fig. 11(b)). In the PEEM imaging experiments, liquid Ga appears bright against a dark GaAs substrate. As can be seen from Fig. 11(a), the solid-liquid interface tends to form contact lines along \(<110>\) crystallographic directions. These straight contact lines frequently transform into curved segments as the droplets etch the substrate and spread laterally. To understand the local energetics, it is therefore of interest to determine the instantaneous contact angle \( \theta \), and Lloyd’s mirror PEEM offers a direct means of reconstructing the geometry of the contact-line dynamics.

![Fig. 11](image)

**Fig. 11** (a) PEEM image of Ga droplets on GaAs (001) at 660 °C. (b) AFM image of the same sample quenched to room temperature \( [6] \) (IOP Publishing).

Figure 12(a) contains a Lloyd’s Mirror PEEM image of part of a rectangular droplet showing Lloyd’s fringes. Experiments are performed using a 100W UV Hg arc-discharge lamp. The electric field used for accelerating the electrons is 20 kV across a 2 mm specimen–anode gap. The angle of the incident photons is \( \alpha = 16^\circ \), with an upper cut-off energy of the Hg arc-discharge lamp approaching 5.0 eV. Since the photoemission threshold for liquid Ga is 4.33 eV \([34]\), the emission is dominated by a single broadened emission line at \( \lambda = 253.65 \) nm with \( \Delta \lambda = 20 \) nm. To a good approximation, the surface profile along the white line in Fig.12(a) can be viewed as 1D. We may therefore use the inverse Hilbert transform (Eq. (8)) to reconstruct the surface shape from the fringe positions. This is achieved via an iterative procedure, giving the interpolated height for the droplet shape function as shown in Fig. 12(b) \([5]\). From this reconstruction, we are able to deduce the contact angle for the Ga droplet as \( \theta = 22^\circ \) at 620 °C, which has important implications for the physical processes governing the local stability of the contact-line \([5]\).

This in situ measurement compares favourably with ex situ non-contact AFM determinations of the contact angle which yield 25 ± 2°. This agreement is perhaps surprising given the quenching, solidification and surface oxidation of the droplets following removal of the sample from the UHV LEEM system. The example illustrated in Fig. 12 is for the reconstruction of a 1D surface profile. However, extension of Lloyd’s fringe measurements to 3D should be entirely feasible via the use of Eq. (7) and this is considered further in \([6]\).
Fig. 12 (a) Lloyd’s Mirror PEEM image of part of a rectangular droplet showing bright Lloyd’s fringes. (b) Surface profile reconstructed using an iterative method based on Eq. (8). The contact angle is determined to be $\theta = 22^\circ$. Eight fringes are included in the iteration. If the two bright fringes closest to the top of the droplet are removed from the data the range of profile variation is indicated by the shaded region which does not influence the contact angle determination [6] (IOP Publishing).

4.4 Extending the resolution of Lloyd’s Mirror PEEM.

Using a simple UV lamp source, the height resolution of the method is limited to $\sim 0.5 \mu m$ (the minimum fringe spacing). This could be considerably improved using high coherence monochromatic synchrotron UV radiation or even soft x-ray PEEM (with elemental selectivity). A plot of critical angle of total external reflection and vertical resolution calculated from Eq. (5) as a function of photon energy is shown in Fig. 13. It can be seen that a resolution of 50 nm is attainable for a photon energy of 50 eV with a critical angle of 15°. Since the method is compatible with existing synchrotron-based XPEEM sites, attaining the improved resolution would seem entirely feasible and open up the possibility of studying surface shapes changes on length scales approaching the nanoscale regime.

Fig. 13 Vertical resolution (solid line) and critical angle of reflection (dashed line) as a function of photon energy [6] (IOP Publishing).

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