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Monitoring surface roughness during film growth using modulated RHEED intensity oscillations

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ABSTRACT

Separation of the high- and low-frequency components of Reflection High-Energy Electron Diffraction (RHEED) intensity oscillations during pulsed deposition allows the extraction of a signal that is in phase with the cyclic surface morphology evolution during layer-by-layer growth. Similar to a biased impedance measurement in electricity, the periodic modulation of surface roughness induced by the pulsed deposition probes the differential response of the growth front to changes in step density. This signal does not follow the complex variation of the RHEED oscillation phase with diffraction conditions and surface reconstruction and therefore allows a direct detection of monolayer completion. Off-Laue Circle oscillations show promise to probe the surface morphology evolution at sharply defined in-plane spatial frequencies.

1. Introduction

Reflection High-Energy Electron Diffraction (RHEED) is a sensitive and versatile tool to monitor epitaxial growth in vacuum or at low pressures [1–3]. In particular, RHEED intensity oscillations [4,5] caused by the periodic surface morphology variations during layer-by-layer (Frank–van-der-Merwe) [6,7] growth provide a convenient and accurate in-situ tool to measure and control layer thickness and composition [8,9].

While the period of the oscillations is directly related to the crystal structure (lattice constant) of the growing material, the phase of RHEED intensity oscillations is a complicated function of the diffraction conditions [10] and the surface reconstruction [11]. Recent theoretical work also indicates that the diffracted intensity depends in a non-trivial way on the fraction of the surface covered by two-dimensional islands (coverage), the areal density of monolayer steps (step density) and even on the orientation of such surface steps, whether they run parallel or perpendicular to the incident beam [12].

As this dependence can be very sensitive (the phase may vary on the order of half a period when changing the incidence angle by as little as 0.1°), it is almost impossible to accurately predict or even reproduce a found correspondence between a position in the periodic growth cycle, such as monolayer completion, and a feature in the intensity oscillations, such as an intensity maximum. This complication rarely plays a role when measuring the growth rate or controlling the layer thickness, as the period can be accurately determined and then extrapolated from the onset of growth [13]. During heteroepitaxy, or when growth instabilities occur, however, we usually cannot decide whether a phase shift in the intensity oscillations is due to a variation in the growth process that affects the surface morphology, or a different mechanism, such as a change in surface reconstruction, that primarily affects the diffraction process.

The question arises: Is there a way to reliably and automatically detect the completion of a monolayer using RHEED despite its complicated diffraction mechanism that almost arbitrarily shifts the phase of the intensity oscillations?

This question is an important one as a dependable characterization of the surface morphology would allow us to better understand growth instabilities and in particular the growth kinetics at heterointerfaces that are so important for the fabrication of heterostructures. Significant progress has been made in this direction by analyzing the recovery of the RHEED intensity after deposition. Early on, it was found [14] that the recovery of the RHEED intensity after growth depends on the position in the layer-by-layer growth cycle. Termination of the growth close to an integer layer coverage led to a fast initial recovery, whereas this recovery is much slower if the growth is terminated at a position in between. The recovery actually consists of two contributions as the initial, fast recovery is followed by a slower process [15]. The two processes can be identified as the initial reduction to a two-level system on the lateral scale responsible for the layer-by-layer oscillations, followed by a larger scale redistribution and smoothing of the surface towards larger scale terraces separated by monolayer steps [16,17].

During pulsed deposition, at least the fast recovery is always present in the intervals between the deposition pulses [18], and the

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recovery time depends on the layer coverage [19]. This has been used in a combined x-ray diffraction and RHEED study to correlate the recovery time between the pulses with the lower frequency intensity variations during the layer-by-layer growth cycle [20]. The fit results confirmed that the variation in recovery time is in phase with the changes in surface roughness and the x-ray intensity, while the RHEED signal can have any phase. The recovery time fits in these experiments are rather tedious to apply, and suffer from relatively high noise levels.

In this paper, we attempt to extract the surface morphology information contained in the RHEED signal of pulsed deposition in a fast and numerically stable way, so that it may be applied in real-time in practical growth situations.

2. Experimental

The growth experiments were performed in a pulsed laser deposition (PLD) system at the Max Planck Institute in Stuttgart studying the homoepitaxy of (001)-oriented strontium titanate ($SrTiO_3$, STO, miscut 0.13°). This system is equipped with a KrF laser (wavelength 248 nm, pulse duration 25 ns) for ablation and a CO_2 laser (9.3 µm wavelength, 1 kW) for direct substrate heating from the back side.

Substrates were prepared by annealing for 5 min at 1200 °C in 5–10 Pa (0.05–0.1 mbar) of molecular oxygen directly prior to growth, eliminating the need for a chemical etching step (termination) prior to loading [21]. Ramp rates were 45 K/min.

Deposition was performed at background pressures of 5.2–5.3 Pa (0.052–0.053 mbar) of molecular oxygen and substrate temperatures of 840 and 1180 °C. The substrate size was 5×5×1 mm^3 and the target–substrate distance was 56 mm. Ablation was performed with a laser fluence of 2.0 J/cm^2 on the target and a pulse frequency of 1 Hz.

RHEED data were acquired using the Safire data acquisition software with a PCO pixelfly USB digital camera running at 36 fps (36 Hz). The RHEED gun was a differentially pumped Staib system operated at 30 keV. A screenshot of the RHEED pattern in false colors directly before the onset of growth is shown in Fig. 1. The azimuth is slightly off a [001] direction, the surface shows a 2 × lateral superperiod. The colored frames mark the regions in which the integrated intensity was measured as a function of time during deposition. A circular line marks the approximate position of the Laue circle, the positions in reciprocal space where the diffraction condition is met in single scattering (kinematical) theory. The big red spot is created by the part of the primary beam that misses the sample, defining the origin of reciprocal space.

The data set used to illustrate and analyze the measurement algorithm is shown in Fig. 2. Grown at 5.2 Pa, the conditions for this layer are deliberately chosen to be oxygen deficient, leading to the eventual collapse of the growth oscillations at about 250 s due to a change in bulk crystal structure [22]. Long before this transition, almost from the beginning of the growth, changes on the surface lead to drastic variations in the amplitude and phase of the oscillations. This, together with the different diffraction conditions for each curve (positions on and off the Laue circle in Fig. 1), represent a wide spread in the different parameters involved in the generation of the RHEED signal, thereby allowing us to probe a large volume of parameter space during the tests.

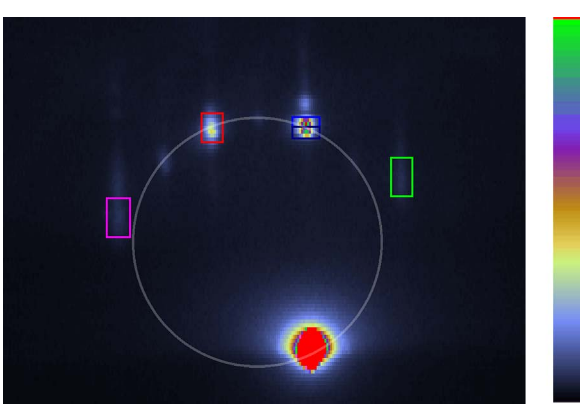

Fig. 1. Screenshot of the RHEED pattern observed during the acquisition of the data for Figs. 2, 5, 6 and 7, taken directly before growth. The averaged intensity in the areas marked by colored frames is measured as a function of time. The approximate position of the Laue circle is indicated by the circular line. The false color bar to the right represents the intensity range of the camera, from zero intensity at the bottom to saturation (red). (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper).

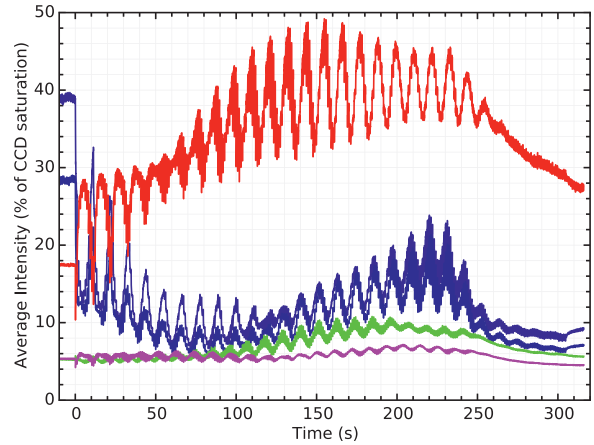

Fig. 2. Data set used to test and validate the processing algorithm. The color of each curve is identical to the frame color of the area it was acquired at in Fig. 1. Substrate temperature 840 °C, oxygen background pressure 5.2 Pa (0.052 mbar). (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper).

3. Concept

The hidden variable. During epitaxy with a constant flux, e.g. from an effusion cell in molecular beam epitaxy (MBE), the atoms or molecules that form the film arrive at a fixed rate. A high crystal quality is often achieved when growing in the layer-by-layer, i.e. Frank–van-der-Merwe growth mode. Under these conditions, two-dimensional islands with a monolayer height defined by the crystal structure nucleate, grow laterally in size and coalesce to form the next completed layer, on which the cycle repeats.

If we plot the evolution of step density versus coverage during this process, we qualitatively obtain the relationship sketched in Fig. 3a). The step density steeply rises as the islands form, reaching a maximum around the percolation threshold when the islands merge, and then plummets as the voids between the islands fill to reach the next minimum when the layer is completed. For ideal layer-by-layer growth, the step density at this minimum is zero, the oscillations are undamped and the growth front never consists of more than two levels. In a real experiment, in particular during the first few oscillations starting from flat terraces, these conditions are often well approximated, and the step density at integer layer coverages can indeed be quite low compared to the largest step density somewhere around half layer coverage, generating a strong contrast. As we observe low damping, we therefore neglect atoms on top of the growing layer, and approximate the coverage as the fractional area of a single growing layer. In continuous growth, we control this coverage by the deposition, and the step density follows, governed by the intrinsic growth kinetics of the surface, to produce the variation that leads to the oscillating RHEED signal.

In a pulsed deposition experiment such as in PLD, the situation is different. With each growth pulse, the coverage abruptly increases by a fixed amount. The step density on the surface dramatically increases at the same time, jumping to a high value. Between the deposition pulses,