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Initial stages of MnAs/GaAs(001) epitaxy studied by RHEED azimuthal scans

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Abstract

We study the nucleation phase of molecular beam epitaxy of (hexagonal) MnAs on (cubic) GaAs (001) using reflection high-energy electron diffraction (RHEED) azimuthal scans. The nucleation proceeds from a non-reconstructed initial stage through randomly oriented small nuclei and two orientation stages to the final single-phase epitaxial orientation. The fascinatingly complex nucleation process contains elements of both Volmer–Weber and Stranski–Krastanov growth. The measurement demonstrates the potential of high-resolution RHEED techniques to assess details of the surface structure during epitaxy.

Keywords: Reflection high-energy electron diffraction (RHEED); Molecular beam epitaxy (MBE); Nucleation; Surface structure; Heteroepitaxy

1. Introduction

The molecular beam epitaxy (MBE) growth of heterointerfaces is usually classified into three different growth modes. In the Frank–van der Merwe mode [1], one monoatomic or monomolecular surface layer is completed before the next one nucleates on top of it, resulting in a flat, planar growth front. On an exactly oriented surface, this growth mode usually leads to a periodically varying surface morphology, resulting in diffraction oscillations [2–4]. In the Volmer–Weber mode [5], the deposited material does not wet the surface and immediately forms disconnected mounds. Stranski–Krastanov growth [6] can be regarded as a combination of the previous two, since it is characterized by a two-dimensional wetting layer followed by the formation of mounds that relax by forming dislocations at the interface. The three categories above are necessarily broad and general, since they operate with the macroscopic concept of wetting. In this work, we attempt to take a closer look at an example of nucleation during heteroepitaxy [MnAs on GaAs(001), [7–9]] to follow the process at the atomic scale. It turns out that although the final film shows good epitaxy and a unique orientation of the film with respect to the substrate, there are several metastable intermediate stages during the nucleation and epitaxy is not established until a nominal layer thickness of 2 ML.

Surface structural investigations are usually conducted using low-energy electron diffraction (LEED). Its normal incidence geometry allows the acquisition of a two-dimensional representation of the surface in a single measurement. On the other hand, a LEED apparatus covers a significant part of the solid angle in front of the sample and is therefore not well compatible with a growth chamber in which this area is occupied by effusion sources. The grazing incidence geometry of reflection high-energy electron diffraction (RHEED) leaves enough room for deposition equipment. Its in-plane diffraction geometry produces cuts through reciprocal space normal to the surface. To access the in-plane geometry of the surface, we therefore rotate the sample. This allows us to obtain in-plane intensity maps similar to LEED.

2. Azimuthal RHEED scans

The reciprocal space geometry of an azimuthal RHEED scan [10] is shown in Fig. 1. Due to the high energy of the

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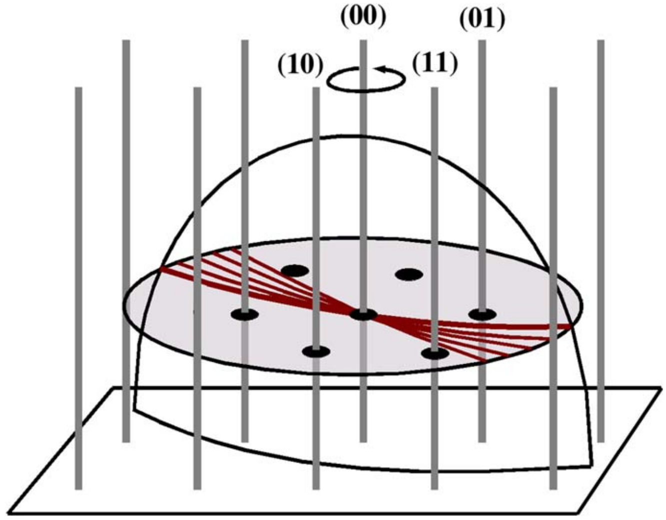

Fig. 1. Geometry of a RHEED azimuthal scan.

electrons, the Ewald sphere is large, resulting in an almost planar almost vertical cut through the reciprocal lattice. During the measurement, the sample is rotated slowly around its surface normal, and the reciprocal lattice moves through the Ewald sphere, projecting the intersection onto the fluorescent screen. To obtain the diffraction pattern in a plane parallel to the surface, the intensities are recorded along a line parallel to the shadow edge at a fixed reciprocal lattice distance l. Within half a rotation, a complete sample of the resulting plane is obtained as indicated by the circular area in Fig. 1. To reconstruct the diffraction pattern, the intensity along the lines just needs to be plotted as a function of azimuthal angle bent approximately with the radius of the Ewald sphere. In contrast to LEED, the surface normal coordinate l in reciprocal space can be chosen independently of the electron energy, and the cut is not spherical but planar. Apart from revealing the full two-dimensional symmetry of the surface at constant l, azimuthal RHEED scans also allow access to the high-resolution direction of RHEED normal to the Ewald sphere. This typically results in a 20-fold improvement of resolution compared to standard RHEED geometries [11].

3. Experiment

The GaAs substrates for the present study were prepared by thermally desorbing the oxide around 580 °C and then growing a GaAs buffer layer with long growth interruptions at approximately 540 °C with a V-III ratio of about 3. The growth rate was two monolayers (ML) per second. These growth conditions are in the center of the $\beta(2 \times 4)$ surface reconstruction stability range for a static (non-growing) surface. The sample was then cooled to 250 °C under the same As flux for the MnAs deposition. The surface exhibited average coherent domain (terrace) sizes of at least 600 nm, determined from the instrument-limited peak width of in situ surface X-ray diffraction [12] before deposition. The surface reconstruction was $c(4 \times 4)$. After doing an initial scan of the bare GaAs surface, we opened the Mn shutter to deposit MnAs continuously while slowly rotating the substrate to acquire the RHEED azimuthal scans. The growth rate was 0.36 ± 0.04 nm/h. It was determined from X-ray reflectivity measurements on thicker layers. Azimuthal scans were acquired at several MnAs coverages ranging from nominal values of 0.03 to 2 monolayers (ML). Since we recorded the measurement line across the complete width of the diffraction pattern, half a rotation was sufficient to reconstruct the complete plane. One half rotation took about 120 s and the acquisition frequency was 25 Hz, resulting in about 3000 lines to build the image. The maximum intensity was adjusted to the peak intensity of the specular spot.

4. Results

The resulting azimuthal scans during the nucleation of MnAs are shown in Figs. 2–7. All scans have the same orientation and size in reciprocal space. Since the RHEED intensities cover a significant dynamic range, the gray scale in the images is made non-linear (Gamma correction) to reveal the weak structures close to the background level. This non-linearity is identical for all patterns except Fig. 7, which is from a different sample. The RHEED intensity was constant except for an increase between Figs. 3 and 4. Fig. 2 shows the GaAs $c(4 \times 4)$ surface just before MnAs deposition. As in the following figures, the $[\bar{1}10]$ direction is upwards. The square surface unit cell with the 2× periodicity along $\langle 110 \rangle$ and 4× along $\langle 100 \rangle$ can be clearly identified and is marked by white circles indicating the (00), (11), (02) and $(\bar{1}1)$ rods at its corners. The area of the scans encompasses more than two diffraction orders of the surface unit cell in both directions. Since the cut is planar and parallel to the surface, the third index l is constant. For all scans except Fig. 4(b) and (c), $l = 0.7 \pm 0.15$. This l position is the geometrical value, without inner potential corrections.

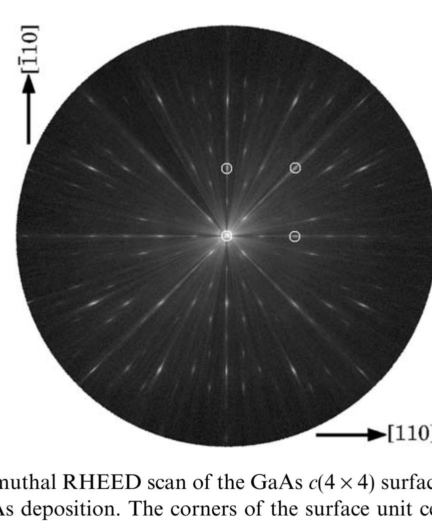

Fig. 2. Azimuthal RHEED scan of the GaAs $c(4 \times 4)$ surface immediately before MnAs deposition. The corners of the surface unit cell are marked by circles.