

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

We have applied the azimuthal reflection high-energy electron diffraction (RHEED) scan technique to study the nucleation and initial growth of MnAs on GaAs(001). The reciprocal space geometry of this measurement is shown in Fig. 34. RHEED intensity appears where the Ewald sphere intersects the reciprocal lattice rods, projected from the center of the Ewald sphere to the screen. The RHEED screen (not shown) is arranged vertically and located parallel to the left edge of the sample surface (black plane) at some distance. For typical RHEED energies, the Ewald sphere is about ten times as large as shown in the image. In an azimuthal scan, the intensity on the screen is measured along a line parallel to the sample surface, for small angles corresponding to the intensity along the recording line in reciprocal space. When we rotate the sample, the Ewald sphere remains stationary, the reciprocal lattice rotates around the (00) rod, and the recording line traces the disc. This plane can therefore be reconstructed by reassembling the recorded lines as a function of azimuthal angle with the known curvature along the Ewald sphere. In this way, a two-dimensional planar cut of reciprocal space parallel to the sample surface is obtained. With sufficiently high acquisition frequency, the evolution of the reciprocal space pattern can be followed in real time during epitaxy.

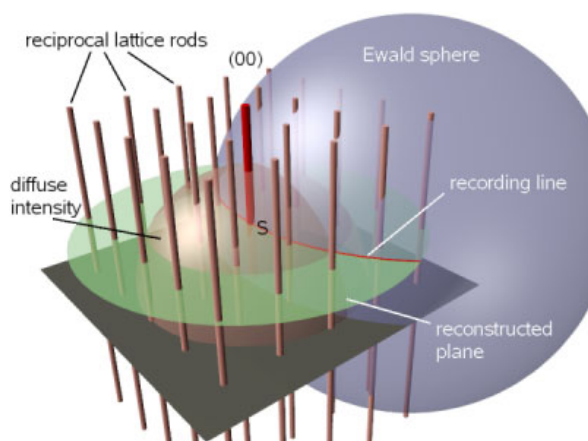


Fig. 34. Reciprocal space geometry of the experiment. During sample rotation, the recording line traces the disc, which then can be reconstructed from the linear intensity as a function of azimuthal angle. The diffuse intensity appears between 0.3 and 0.7 ML coverage.

Following GaAs buffer growth under standard conditions of molecular-beam epitaxy (MBE), the surface was cooled to 250 °C. On the resulting  $c(4\times 4)$  reconstruction [cf. Fig. 35(a)], we deposited MnAs at a rate of  $0.36 \pm 0.04$  nm/h. During the deposition of the first nominal 0.25 monolayers (MLs) of MnAs, the surface reconstruction disappeared without a significant increase in background intensity [Fig. 35(b)]. We interpret this as an adsorption of the Mn at randomly occupied regular positions within the fundamental  $1\times 1$  unit cell, thereby not increasing the disorder (diffuse intensity). From 0.3 to 0.7 ML nominal coverage, the diffuse background was stronger, and a spherical shell of diffuse intensity appeared [cf. Figs. 35(c)–35(e)]. Its radius is around 2.5 Å, in agreement with typical nearest-neighbor distances in MnAs. To verify the spherical shape, reciprocal space planes at different distances from the surface were processed as shown in Figs. 35(d) and 35(e). The same 2.5 Å distance also appeared in in-plane reflections marked by the circles. Obviously, microscopically small units with random orientation and MnAs crystal spacing are formed. It is likely that either part of such a unit or the whole unit itself is mobile, only weakly interacting with the periodic potential of the surface. The marked reflec-

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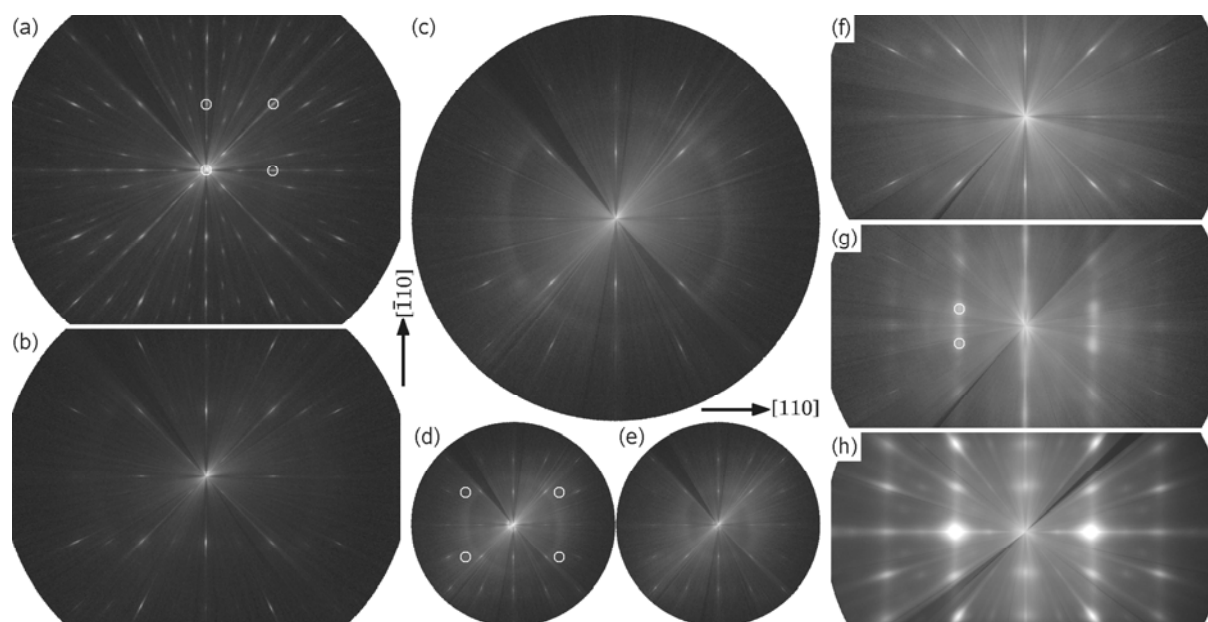


Fig. 35. RHEED azimuthal scans before and during the deposition of MnAs on GaAs(001). In (a), the unit cell of the initial  $c(4 \times 4)$  surface is indicated by circles. The different stages are: (b) removal of the reconstruction at 0–0.25 ML, (c)–(e) appearance of a spherical shell of diffuse intensity at 0.3–0.7 ML, (f) increased diffuse background at 0.8–1.2 ML, (g) epitaxial lock-in along  $[110]$  around 1.8 ML, (h) full 2D epitaxy at 6.4 ML. Planes (d) and (e) were recorded together with (c), but at higher positions along  $(00)$  to verify the spherical shape shown in Fig. 34.

tions indicate a weak preferential alignment, where obviously the  $2.5 \text{ \AA}$  distance finds a weak local potential minimum.

Very soon after this orientationally random state, at coverages between 0.8 and 1.2 ML [Fig. 35(f)], a mostly featureless diffraction pattern with an intense diffuse background was observed. The density of MnAs on the surface was now sufficiently high so that the interaction with the substrate could no longer dominate its behavior, and steric effects played a significant role. At 1.8 ML nominal coverage, the bonds within the MnAs became sufficiently ordered to allow for a preliminary epitaxial lock-in to the substrate. This took place first along the  $[110]$  direction, with narrow reflection shapes, while the reflections were still quite broad along  $[\bar{1}10]$ . The formation of the interface proceeded via intermediate periodicities marked by the circles in Fig. 35(g).

These intermediate reflections vanished suddenly around 2 ML coverage (not shown), when the final symmetry shown in Fig. 35(h) was established and the full epitaxial order in both directions was obtained. The intermediate reflections of Fig. 35(g) are half way in reciprocal space to the reflections of the final pattern, which means that the epitaxial lock-in proceeded through an intermediate state with twice the period of the final configuration in real space. Further growth did not change the symmetry of this pattern, only the intensity of the reflections increased as the surface became ordered and smoother.

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