Growth control of MnAs on GaAs(001) by reflection high-energy electron diffraction

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The MnAs($\bar{1}100$) surface is investigated during growth by reflection high-energy electron diffraction (RHEED). (1×2), (1×1), (2×1) and (4×1) RHEED patterns have been observed by varying the growth conditions, indicating various stoichiometry dependent reconstructions. A phase diagram showing the dependence of the reconstructions on the growth parameters is presented. RHEED intensity oscillations have been found, evidencing layer-by-layer growth of MnAs. © *2000 American Institute of Physics.* [S0003-6951(00)02107-0]

The growth of magnetic materials on semiconductors attracts considerable attention due to the potential of integrating magnetism with semiconductor electronics.[1] In view of the desired interface and thickness control, molecular-beam epitaxy (MBE) provides an elegant way of fabricating such ferromagnetic-semiconductor hybrid structures. Recent investigations have shown that MnAs is a promising candidate from the epitaxial point of view. In spite of the large misfit, MnAs grows well oriented on GaAs(001) and forms a stable ferromagnetic layer with good structural properties[2,3]. In order to optimize the layer and interface quality it is essential to control the surface during growth by *in situ* techniques, in particular, by reflection high-energy electron diffraction (RHEED) as a standard method in MBE. The growth mode and surface reconstruction play an important role, especially for growing heterostructures. Recently, it has been shown for MnAs/GaAs/MnAs trilayer heterostructures on GaAs(111)B that GaAs grows monocrystalline on the As-rich MnAs(0001)-(3×2) surface but polycrystalline on the MnAs(0001)-(2×2) surface.[4] In this letter, we present a systematic study of the dependence of the surface reconstructions on the growth conditions for MnAs($\bar{1}100$), which is the relevant orientation for heterostructures grown on GaAs(001). In addition, we demonstrate by recording of RHEED intensity oscillations that a layer-by-layer growth can be realized and that the layer thickness is well controlled.

MnAs was grown on GaAs(001) substrate by using standard MBE with an As valve cracker cell, which allows to change the beam-equivalent-pressure (BEP) of As_4 within two seconds over one order of magnitude. After growing a 100 nm thick buffer layer of GaAs at a substrate temperature $T_s = 550\,^\circ$C, the sample was cooled down to 250 °C, leading to the GaAs(001)-c(4×4) reconstruction. On this As-rich template, we have grown MnAs with a growth rate of 10 nm h^{-1} at $T_s = 250\,^\circ$C. During the MnAs growth the as-measured As_4/Mn BEP ratio was fixed at 350. The epitaxial orientation with respect to the GaAs substrate was found to be: MnAs($\bar{1}100$)||GaAs(001) and MnAs[0001]||GaAs[$\bar{1}10$] (type A orientation[2,3]). A 30 nm thick MnAs film was grown in order to have a MnAs template for studying different surface reconstructions. The RHEED patterns were recorded by an image acquisition system which consists of a CCD camera connected to a computer.

The substrate temperature was varied within a range from 250 up to 600 °C and the As_4/Mn BEP ratio between 80 and 480, respectively. This parameter set covers most experiments concerning the usual MnAs growth by MBE. The whole parameter range was scanned by starting from $T_s = 250\,^\circ$C and an As_4/Mn BEP ratio of 480.

At typical growth conditions the MnAs($\bar{1}100$) surface is (1×2) reconstructed. The corresponding RHEED patterns are shown in Fig. 1(a). This surface configuration is observed up to 350 °C for an As_4 BEP of 7.2×10^{-6} Torr and up to 330 °C for 1.5×10^{-8} Torr, respectively. For higher substrate temperatures, the (1×2) reconstruction changes to a (1×1) unreconstructed surface, because As evaporates from the surface. The interplanar spacings of 5.7 and 3.7 Å, extracted from the (1×1) RHEED pattern shown in Fig. 1(b), correspond to the surface unit mesh of the bulk structure. Further increase of T_s results in a Mn-rich surface of MnAs. At first, we observe a (2×1) RHEED pattern as shown in Fig. 1(c). At even higher temperatures, the (2×1) reconstruction changes to a (4×1) structure, as shown in Fig. 1(d). The intensity of the quarter-order streaks in the [$11\bar{2}0$] azimuth increases gradually with temperature, in contrast to a rather abrupt intensity change of the fractional-order streaks for the other phase transitions. This suggests the coexistence of the (2×1) and (4×1) reconstructed domains or a rather disordered (4×1) structure over an extended temperature range. As a result, we find that the ($11\bar{2}0$) surface of MnAs epitaxially grown on GaAs(001) forms four different types of stable surfaces in dependence on the surface stoichiometry:

(i) As-rich with (1×2) structure,
(ii) transition range with (1×1) structure,
(iii) Mn-rich with (2×1) structure, and
(iv) Mn-rich with (4×1) structure.

Higher substrate temperatures than 580 °C at an As_4/Mn BEP ratio of 80 and 600 °C at an As_4/Mn BEP ratio of 480 favor the evaporation of As from the surface and the growth front becomes too Mn-rich which results in surface damage and roughening.

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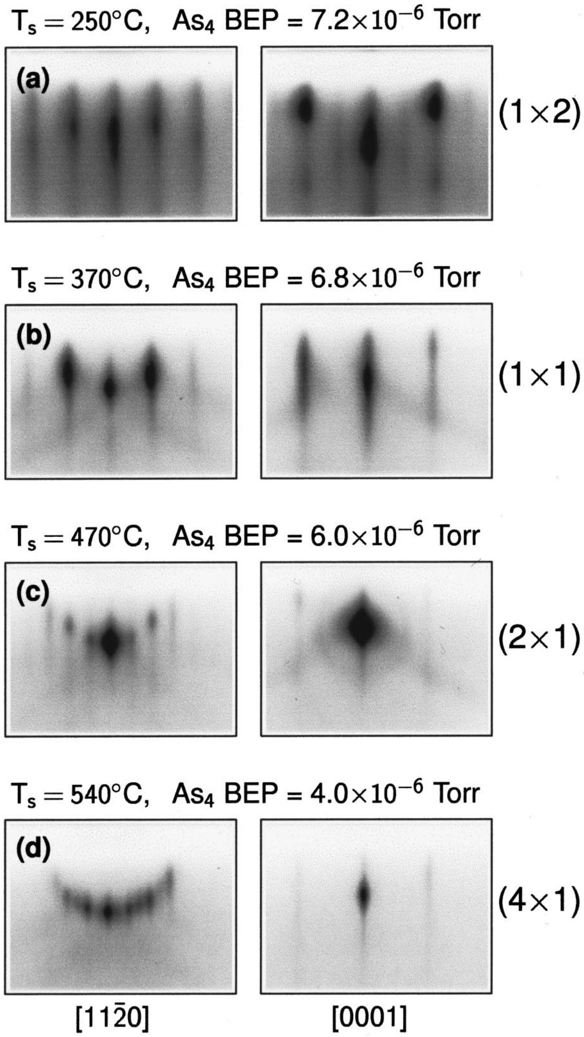

FIG. 1. Characteristic RHEED patterns of MnAs($\bar{1}$100) along the [11$\bar{2}$0] azimuth (left panels) and [0001] azimuth (right panels) recorded during growth at different substrate temperatures and As_4 BEP values. MnAs growth rate: 10 nm h^{-1}.

Figure 2 shows the surface phase diagram resulting from our systematic RHEED investigations. It has been constructed from 600 RHEED patterns of the MnAs($\bar{1}$100) surface, obtained by scanning the temperature at a constant As_4/Mn BEP ratio and by scanning the As_4/Mn BEP ratio at constant temperature. It is important to note that within the accuracy of the measuring procedure, no hysteresis effects have been observed, i.e., a change of reconstruction was accomplished within a few seconds.

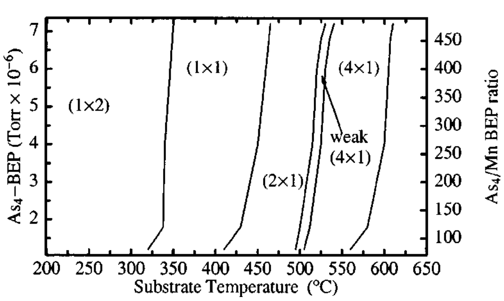

FIG. 2. Surface phase diagram for MnAs($\bar{1}$100) growth from Mn and As_4 molecular beams. MnAs growth rate: 10 nm h^{-1}.

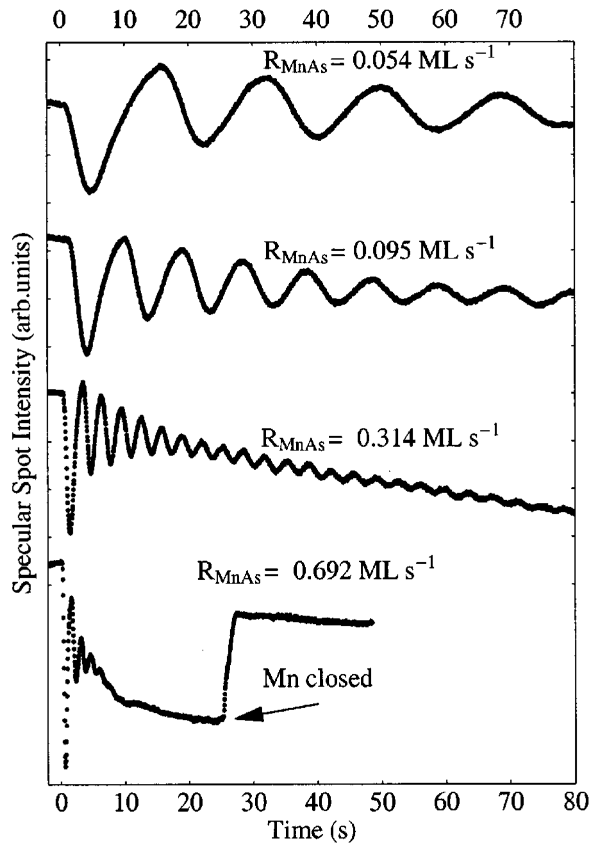

FIG. 3. RHEED intensity oscillations of the specular spot measured in the MnAs[11$\bar{2}$0] azimuth as a function of growth rate R_{MnAs}. The surface was grown at a substrate temperature of 265 °C after a short annealing at 300 °C. The Mn flux was applied at $t=0$.

Additionally, we have been able to record RHEED-intensity oscillations during MnAs growth. The time dependence of the intensities for different growth rates is shown in Fig. 3. The recording was performed by measuring the intensity of the specular spot in the MnAs[11$\bar{2}$0] azimuth. Prior to the oscillation measurement the surface was annealed for 2 min at 300 °C. This remarkably improves the surface quality as reported in earlier work.[5] It should be noted that for lower growth rates $R_{MnAs} \leqslant 0.54$ ML (monolayers)s^{-1} the intensity did not increase after closing the Mn shutter, while for $R_{MnAs}=0.65$ ML s^{-1}, a sudden increase is observed. This indicates that at lower growth rates the surface is closer to the thermal equilibrium. Higher rates result in a kinetically determined growth mode with an improving surface (ordering and smoothing) after the growth has been stopped. Altogether, these results evidence that MnAs can be grown in the layer-by-layer mode which is the desired mode for preparing smooth epitaxial films and interfaces.

In conclusion, we have presented a systematic study of surface reconstructions of MBE-grown MnAs on GaAs(001), summarized in a surface phase diagram. The existence of various stoichiometry dependent reconstructions during growth of the MnAs film on the very dissimilar GaAs substrate, concerning structure and properties, indicate the growth of a well-ordered material. This is confirmed by observing RHEED intensity oscillations. The weak dependence of the reconstructions on the As_4/Mn BEP ratio allows a stable MnAs growth within a wide parameter range.

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