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Atomic scale morphology of self-organized periodic elastic domains in epitaxial ferromagnetic MnAs films

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The surface of epitaxial MnAs layers grown on GaAs(001) substrates by molecular beam epitaxy is studied by atomic force microscopy and scanning tunneling microscopy (STM). A periodic array of alternating ridges and grooves is observed. The periodicity ranges from 200 to 900 nm and increases with increasing layer thickness. The terrace-step morphology and the surface reconstruction on the ridges and in the grooves are imaged by STM. It is found that both are independent from the ridge-groove structure, supporting the idea that the formation of the ridge-groove structure is due to elastic distortion of the film during cooling after growth. © *2002 American Institute of Physics.*
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INTRODUCTION

MnAs layers epitaxially grown on GaAs(001) may separate into two distinct phases.[1,2] This behavior is particularly intriguing because one phase is ferromagnetic, while the other is paramagnetic. The two phases arrange in a regular pattern, and thus heteroepitaxial growth of MnAs provides an easy and inexpensive way of fabricating magnetic nanostrucures on GaAs.

For bulk MnAs only one phase exists at a given temperature. In strained films, however, the hexagonal ferromagnetic α phase and the orthorhombic paramagnetic β phase coexist near the temperature of the bulk phase transition at $\sim 40\,°C$.[1] The phase composition depends on temperature: In the coexistence region the fraction of β-MnAs increases almost linearly with increasing temperature, until finally the film consists completely of β-MnAs (above roughly 45 °C).[1] A self-organization mechanism separates the coexisting phases into a periodic array of stripes along the MnAs[0001] direction.[2–4] The explanation of this mechanism is based on the elastic strain and on the difference in lattice constants of α and β phase. The requirement that the elastic energy is minimized at equilibrium determines the fraction of β-MnAs in the film and the size of the elastic domains. Model calculations predict that the periodicity of the stripe array scales linearly with the thickness of the MnAs layer.[3] Magnetic force microscopy reveals that the stripes indeed have different magnetic properties.[2] Atomic force microscopy (AFM)[2] and temperature dependent scanning probe microscopy[4] show a periodic height modulation of the morphology, with a periodicity that changes as the amount of the β phase in the film changes with temperature. Thus, there is a large amount of experimental evidence supporting the elastic model. However, direct images of the stripe structure with atomic scale resolution are still lacking. Scanning tunneling microscopy (STM) is an ideal tool to fill this gap.

Here we present AFM and STM images of the stripe structure of epitaxially grown MnAs films on GaAs(001). The images show that the terrace-step-morphology is independent of the stripes. This provides additional evidence that the stripe pattern is due to elastic distortion during cooling after growth of the film, rather than due to kinetic effects during growth.

EXPERIMENT

The MnAs layers were grown by molecular beam epitaxy (MBE) on commercially available epiready GaAs(001) substrates using an MBE system as described previously.[5] MnAs layers were grown on a 100 nm GaAs buffer layer. Substrates and buffer layers were Si doped because conductive samples are necessary to permit STM imaging. MnAs is already conductive[6] because it is a semimetal. MnAs films were grown at substrate temperatures ranging from 200 to 250 °C with a rate of 20 nm/h and an As_4/Mn beam equivalent pressure ratio of 250. The temperature was measured by a thermocouple and the cell fluxes were calibrated by reflection high energy electron diffraction intensity oscillations.[7,8]

For STM investigations we employed a rapid thermal quenching once the growth stoped, in order to conserve the growth morphology during cooling. All impinging fluxes and the substrate heater were switched off, and the sample was quickly removed from the growth chamber, usually within less than 1 min. Then the sample was transferred through ultrahigh vaccum (UHV) to the STM chamber. STM images were taken in UHV using a standard Park VP2 STM.

For the AFM images shown here cooling was performed such that the highest possible uniformity of the stripe structure was achieved. In this case, we cooled the samples to 200 °C at a cooling rate of 20 °C/min and then continued at a rate of 1 °C/min down to room temperature, thus passing the phase transition (around 40 °C) at a very low cooling rate. AFM images were taken *ex situ* in ambient air. Finally, the MnAs layer thickness was routinely measured by scanning electron microscopy of the sample cross section.

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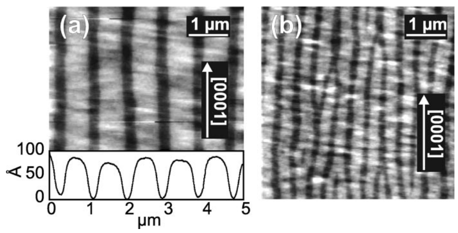

FIG. 1. AFM images of MnAs layers grown at 250 °C, with a layer thickness of: (a) 130 nm and (b) 95 nm. The periodic height modulation is due to a coexistence of α-MnAs (bright) and β-MnAs (dark) in the strained film.

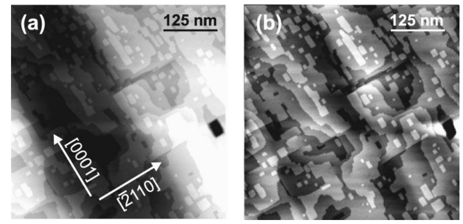

FIG. 3. STM image of a 130 nm thick MnAs film grown at 250 °C. (a) The bright region is α-MnAs, the dark region is β-MnAs. Monolayer steps are also resolved. (b) Same raw data as in (a), after removal of long wavelength undulations by image-processing for a better display of the step morphology.

RESULTS AND DISCUSSION

Figure 1(a) shows an AFM image of a 130 nm thick MnAs layer grown at 250 °C. A line scan showing the cross-sectional profile of the layer is also included. A periodic array of ridges and grooves is visible, the direction of the grooves being along the MnAs[0001] direction (for a description of the epitaxial relationship between MnAs and GaAs see, e.g., Refs. 9–12). The amplitude of the height modulation is 80 Å and the periodicity is 880 nm. Because α-MnAs has the larger lattice constant perpendicular to the film, the ridges in this structure consist of hexagonal α-MnAs, and the grooves consist of orthorhombic β-MnAs. The same structure is observed in thinner MnAs layers, but the periodicity and amplitude of the modulation are reduced. Figure 1(b) shows an AFM image of a 95 nm thick MnAs layer. The amplitude of the height modulation is 50 Å and the periodicity is 400 nm. The elongated features perpendicular to the grooves are due to a small-scale morphology which is not resolved in detail by AFM. In Fig. 2 the periodicity of the ridge-groove structure is plotted as a function of the MnAs layer thickness. The increase of the periodicity with increasing layer thickness is evident.

Figure 3(a) shows a STM image of a 130 nm thick MnAs layer grown at 250 °C. The dark region running from the upper left corner of the image downwards to the right is a groove consisting of β-MnAs. Monolayer steps are also resolved in the image, but the contrast is dominated by the strong height modulation due to the ridge-groove structure. Figure 3(b) shows the same raw data again after different image processing. The long wavelength undulations of the ridge-groove structure are removed by a suitable background subtraction procedure. The terrace-step-morphology remains and is more clearly visible. Steps are predominantly straight and run along the [0001] and [$\bar{2}$110] directions of MnAs, but some rounded step segments are also present. This finding indicates that the growth temperature was high enough to permit the formation of kinks to some extent. A large number of small rectangular islands is observed on the terraces. This observation shows that the temperature is yet not sufficiently high for step flow growth to take place. Comparison of Figs. 3(a) and 3(b) shows that the step morphology is uniform everywhere in the image, in particular it is identical for ridges and grooves. Thus, the growth morphology is independent of the ridge-groove structure. The image shows that the ridge-groove structure is superimposed on the terrace-step morphology, and the surface including its step-morphology is buckled as a whole. In fact, this is what we would expect from the previously mentioned elastic model.[1] The step morphology forms during growth at elevated temperature, when the film consists of pure β-MnAs.[1] On the other hand, the ridge groove structure evolves during cooling when the α-phase forms (around 40 °C). Changes in the step morphology are then no longer possible because this temperature is too low for significant mass transport by diffusion. The terrace-step-morphology should thus be independent from the ridge-groove structure, just as observed.

For comparison, Fig. 4 shows STM images of a 130 nm

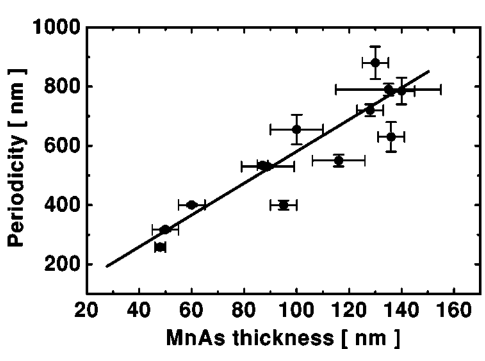

FIG. 2. Periodicity of the ridge-groove structure as a function of the thickness of the MnAs layer.

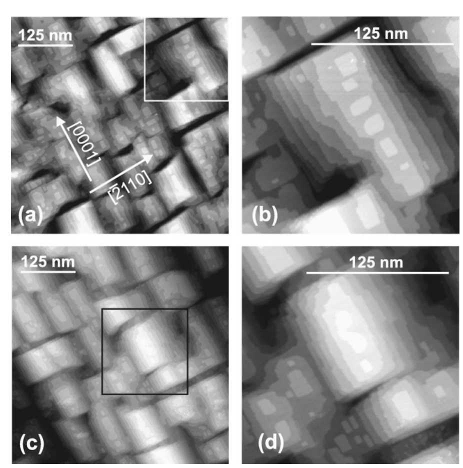

FIG. 4. STM image of a 130 nm thick MnAs film grown at 200 °C. (a) and (c) show images of different portions of the surface of the same film. The roughness is increased (cf. Fig. 3) and bunching of steps along the [0001] direction leads to characteristic features as shown in the magnified parts in (b) and (d).

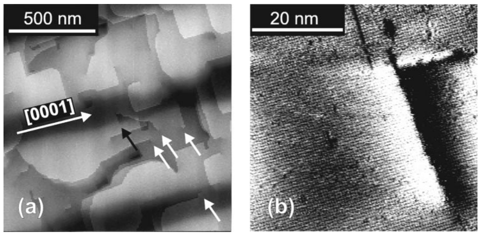

FIG. 5. STM images of a MnAs film with 90 nm thickness grown at 230 °C, after annealing at 300 °C. (a) Rectangular islands as in Fig. 3 are removed by annealing. The arrows mark the positions of dislocations threading to the surface. The region around the dislocation that is marked by the black arrow is magnified in (b), where the (1×2) surface reconstruction is also resolved.

thick MnAs layer grown at a lower substrate temperature of 200 °C. Here, the density of monolayer-steps, i.e., the roughness, is higher than in Fig. 3, and the steps are more straight. There is also a pronounced bunching of steps with edges along the $[\bar{2}110]$ direction. In contrast, the steps with edges along the [0001] direction are well separated. This leads to a characteristic structure of small pyramids with steep facets towards the [0001] directions and shallow facets toward the $[\bar{2}110]$ directions, as shown in detail in the closeups of Figs. 4(b) and 4(d).

Figure 5 shows two STM images of a 90 nm thick MnAs layer grown at 230 °C. Figure 5(a) is a large scale image, showing the elastically determined ridge-groove structure and the step morphology of the surface. The sample shown in this image was annealed after growth for 8 min at 300 °C. Upon annealing, small rectangular islands like those observed in Fig. 3 disappear, and the terrace width increases. Furthermore, the fraction of rounded step edges becomes larger. Screw dislocations generating steps are observed on the ridges as well as in the grooves; some examples are marked by arrows in Fig. 5(a). The screw dislocation marked by the black arrow in Fig. 5(a) is located near a boundary between α and β phases. The threading point lies on a ridge. The step edge associated with the dislocation runs along the $[\bar{2}110]$ direction into a groove, where it has a kink and continues along the [0001] direction. Figure 5(b) shows the step edge from the threading point to the kink with higher resolution. The MnAs$(\bar{1}100)-(1\times2)$ reconstruction[8,10,13] of the surface is clearly resolved. The height modulation due to the transition from the α-MnAs ridge to the β-MnAs groove is again not visible because a linewise background subtraction was performed in order to display the small corrugation of the surface reconstruction. However, from the image in Fig. 5(a) one can see that the lower part of Fig. 5(b) (where the threading point is) shows the surface on the ridge, while the upper part (where the kink is) shows the surface in the groove. The (1×2) reconstruction is observed on the ridge as well as in the groove. Although there is a difference in symmetry between the (hexagonal) α-phase and the (orthorhombic) β-phase, the crystal structures are related.[14] Therefore it seems reasonable that also similar surface structures form.

In summary, we have imaged the morphology and the structure of an array of self organized periodic elastic domains in epitaxial MnAs films on GaAs(001). Two contributions to the morphology were observed: (i) periodic undulations with a period of 200–900 nm and (ii) the step morphology and the atomic structure of the surface. The former is caused by the elastic distortion of the film during cooling due to the presence of α- and β-phase MnAs in the film; the latter is formed during growth. Both are independent from each other. These findings provide further evidence that the ridge-groove structure is due to elastical distortion of the film after growth (during cooling) rather than due to a kinetic growth effect.

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