Effect of strain on the local phase transition temperature of MnAs/GaAs(001)

J. Mohanty, T. Hesjedal,[a] A. Ney, Y. Takagaki, R. Koch, L. Däweritz, and K. H. Ploog
Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5-7, D-10117 Berlin, Germany

(Received 10 June 2003; accepted 13 August 2003)

We present measurements of the influence of local strain on the phase transition behavior of epitaxial MnAs films on GaAs(001). As shown previously, stripes of ferromagnetic α-MnAs and paramagnetic β-MnAs coexist around room temperature. Temperature-dependent atomic force and magnetic force microscopy reveals that the characteristic temperature T^*, at which the as-grown films transform to the paramagnetic β-phase, is locally shifted up towards the value of unstrained bulk MnAs. The film areas exhibiting a higher T^* were identified as regions in which the strain in the MnAs film was allowed to relax.
[DOI: 10.1063/1.1615836]

During the 1990s, the increase of the storage density of magnetic disks outpaced even its electronic counterpart, the number of transistors per integrated circuit, known as Moore's Law.[1] To further increase the density of magnetically based data storage solutions, among other tasks, the efficient production of magnetic nanostructures has to be achieved. For this purpose, self-organized preparation techniques that do not depend on costly nanofabrication schemes may play an important role. On the other hand, in all micro- and nano-structured systems, the physical properties, in particular the magnetic ones, depend on the altered elastic environment.[2]

MnAs is one of the few ferromagnetic metals that can be grown epitaxially on different GaAs planes by solid-source molecular-beam epitaxy.[3,4] On GaAs(001), As-rich growth conditions predominantly lead to the so-called A-orientation with MnAs$(\bar{1}100)\|$GaAs(001) and MnAs[0001] (c-axis)$\|$GaAs$[1\bar{1}0]$. At T_c, just above 40 °C, bulk MnAs exhibits a first-order phase transition between the hexagonal ferromagnetic α-MnAs phase and the orthorhombic paramagnetic β-MnAs phase, which is contracted in volume by 2% (Ref. 5). In thin MnAs films on GaAs(001), on the other hand, the two phases coexist over a temperature range from 10 to 40 °C due to the involved strain.[6,7] They form an array of periodic α-/β-MnAs stripes along the MnAs[0001] direction. We define the characteristic transition temperature T^* as the temperature at which the magnetic contrast, as observed by magnetic force microscopy (MFM), vanishes. Above T^*, α-MnAs is completely transformed into paramagnetic β-MnAs.

In this letter, we study the local effect of strain on the structural and magnetic properties of MnAs on GaAs(001). By performing temperature-dependent measurements of the topography and the magnetic contrast by scanning force microscopy, the influence of local strain on the transition temperature T^* was imaged with high lateral resolution from 20 to 45 °C.

Figure 1(a) shows the topography of the α-/β-MnAs ridge–groove structure of a 215-nm-thick MnAs film. The bright structures are α-MnAs extending in height by roughly 1.9% of the film thickness above the dark β-MnAs stripes. The details of the magnetic properties and the stress of MnAs films in the phase transition region were studied recently with variable-temperature MFM[8] and by a cantilever beam technique.[9]

We used a wet chemical etch solution to open up a 60 $\times 60$ μm^2 window in the MnAs surface (see sketch in Fig. 1).[10] This process was introduced recently for the fabrication of regular arrays of MnAs nano-islands.[11] Upon etching at room temperature, already at the early stages the strain is relieved by the formation of cracks running solely along the $[11\bar{2}0]$ direction.[12] These cracks usually exhibit a regular distance from each other and laterally penetrate deep under the resist defining the etch window. Figure 1(b) shows an atomic force microscope (AFM) scan of the MnAs surface close to the etch window. Higher magnification images [Fig. 1(c); position indicated in 1(b)] reveal very narrow cracks with a width beyond the resolution limit of the instrument. Corresponding line scans below and on the right-hand side of the image provide further insight into the crack structure. First, the regular ridge–groove stripe pattern is decoupled across the crack. MFM images (not shown) further reveal that there is no magnetic domain correlation within a stripe across the crack. Second, the line scan along an α-MnAs stripe shows that the film bulges out at the position of the crack. The width of the distortion extends over roughly 1 μm and leads to an increase in height by about 4 nm. The etching process leading to the cracks takes place by a preferential dissolution of the Mn atoms, thus accounting for the directionality in the $[11\bar{2}0]$ MnAs direction (that is, along the hexagonal Mn planes), leaving amorphous As clusters behind.[12] Both processes together, that is, the directional etching and the inherent stress in the film, lead to the formation of a regular array of cracks with an average distance (of a fully cracked film) of roughly the α-/β-stripe periodicity of 1.2 μm.

Upon heating of the MnAs film, the amount of α-MnAs decreases steadily until the whole film is in the β-phase above the transition temperature T^* (uncracked film) of

[a]Author to whom correspondence should be addressed; electronic mail: hesjedal@pdi-berlin.de

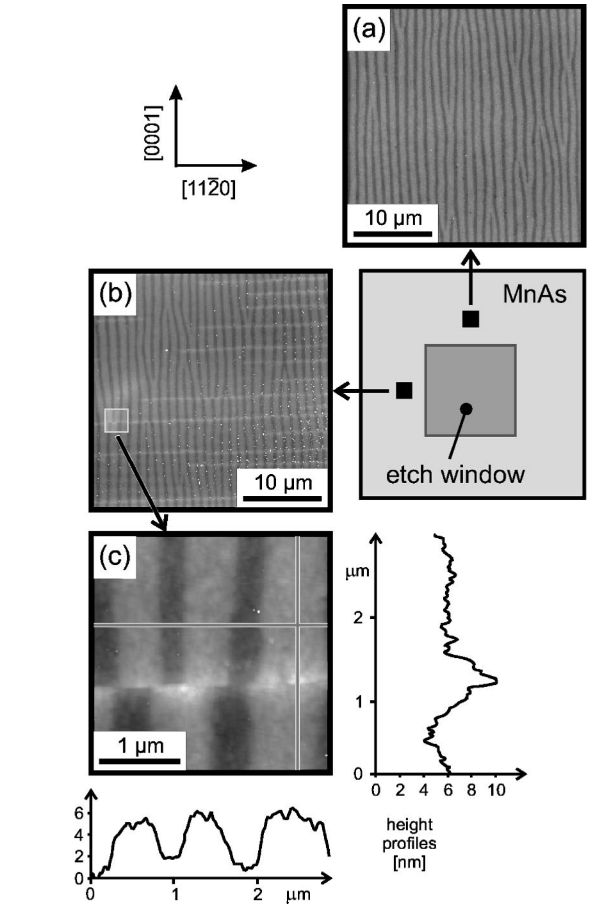

FIG. 1. 30×30 μm^2 AFM topography images of the uncracked (a) and cracked (b) MnAs surface. The 215-nm-thick MnAs film was wet etched through a window (see dark gray area) leading to a regular row of cracks running in the [11$\bar{2}$0] direction. The zoom of the cracked surface area (c) shows a shift of the periodic α-/β-MnAs stripe structure across the crack. The line scan across the crack reveals that MnAs is piling up upon crack formation.

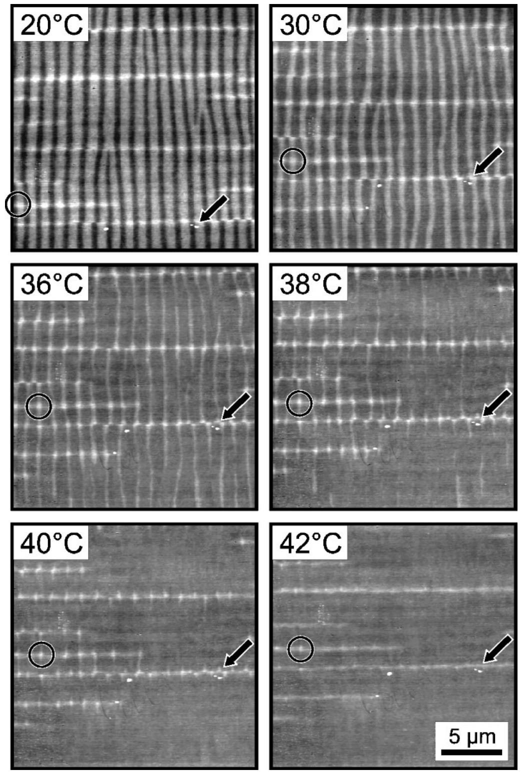

FIG. 2. 20×20 μm^2 topography scans. Evolution of the α-/β-MnAs stripe structure upon heating from 20 to 42 °C. With increasing temperature, the amount of α-MnAs decreases, as can be seen by the decreasing width and length of the bright stripes. Above 40 °C, almost all the entire α-phase has vanished, except in the vicinity of the cracks.

around 38 °C. In the final stage of the transition, that is, a few degrees below T^*, the stripes transform into separated ferromagnetic dots.[8] In Fig. 2, representative topography scans (20×20 μm^2) of the heating sequence ranging from 20 to 42 °C are shown. Circles and arrows indicate the same spots on the film as the scan window is drifting upon heating. From 20 °C to above 30 °C, the α-stripes become narrower; however, the stripes are still continuous, forming a regular array. Around 36 °C, the stripes decompose into smaller segments that extend from the cracks. Above 40 °C, the stripe segments shrink into isolated features that are pinned by the crack line and persist even at 42 °C. Looking in more detail at the cracks in this temperature range reveals that the surface appearance of the cracks, that is, their width and length, is not affected by thermal cycling.

To investigate the stripe configuration in more detail, we performed comparative MFM measurements at 20 and 37 °C; that is, just below T^*. Figure 3 shows 7×3.5 μm^2 images in two different regions of the sample: a cracked and an uncracked area, as indicated in Fig. 1. The upper half-images show the topography of the respective region and the lower half-images the corresponding magnetic contrast. At 20 °C, the α-/β-stripe structure still is predominant, cracks running perpendicular to the stripes are clearly visible in Fig. 3(a). The magnetic contrast displays complex features on the ferromagnetic α-stripes. The easy axis of magnetization is the [11$\bar{2}$0] direction of MnAs; that is, the in-plane direction perpendicular to the stripe orientation.[13] Since the tip of the MFM is magnetized along its axis, the contrast in the MFM images is predominantly due to out-of-plane magnetic field components of the sample. Details of the origin of the magnetic contrast are discussed in more detail elsewhere.[8] For this investigation, it is only of interest that the domain pat-

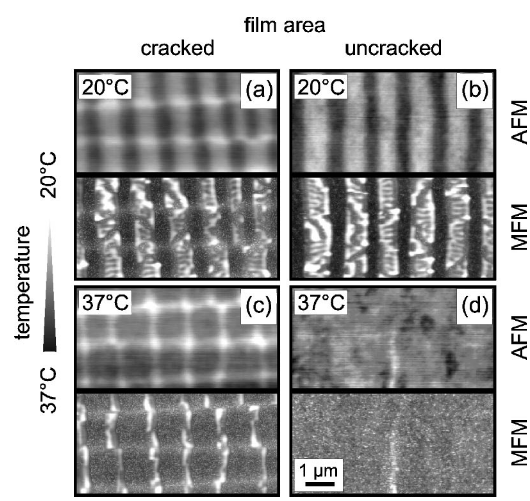

FIG. 3. AFM (upper half) and MFM (lower half) scans of a cracked (left column) and an uncracked (right column) surface area, as described in Fig. 1. The upper row images were taken at 20 °C and the lower row images at 37 °C; that is, close to the phase transition temperature.

terns in the cracked and the uncracked area show no significant differences well below T^*.

At 37 °C, just below T^* of this particular as-grown film, the topography as well as the magnetic scans show surprising differences. In the topography of the uncracked film [Fig. 1(a)], hardly any α-stripes are visible. In the cracked film [Fig. 1(b)], on the other hand, both the stripe structure as well as the perpendicularly oriented cracks are still present. The crossing points of both patterns stick about 6 nm out of the surrounding β-phase material, giving rise to a two-dimensional (2D) network of protrusions. As found recently, this array can be etched down to the bare substrate, leaving a perfectly ordered pattern of 2D units behind.[11] The corresponding magnetic measurements at 37 °C show almost no contrast on the uncracked surface. In case of the cracked surface area, the α-stripes narrow with increasing temperature and the magnetic domain configuration is less complex compared to 20 °C. The in-plane domains extend over the whole stripes in the $[11\bar{2}0]$ direction and lead, depending on the parallel or antiparallel alignment of the moments, to segments of bright contrast on either edge of the stripes [see Fig. 3(c)]. This contrast is due to the stray field at the edges of the bar-magnet-like domains, pointing up or down depending on the orientation of the in-plane magnetic moments. The 180° domain walls of Bloch-type are visible between the antiparallel aligned domains.

The comparison of the AFM and MFM scans at 20 and 37 °C leads to the conclusion that the α-phase survives in the vicinity of the cracks at elevated temperatures (see Fig. 2, 40 °C), whereas it has disappeared in the uncracked film. According to the MFM scans, remains of the stripes consist of ferromagnetic α-MnAs with T^* being larger in the vicinity of the cracks. From measurements at different temperatures, we estimate a local increase of T^* of the order of 5 °C for these regions; that is, almost reaching the α-/β-phase transition temperature T_c of bulk MnAs.

The shift of the transition temperature T^* in the vicinity of the cracks can be correlated with the local change of the strain state of the film. According to previous studies of bulk MnAs, the transition temperature T_c decreases under pressure,[14] that is, with increasing compressive strain, which was attributed to a strain-dependent magnetic exchange energy.[15] Whereas the misfit strain is relaxed to a large extend at the preparation temperature of 250 °C (Ref. 4), a tensile strain arises upon cooling to room temperature since the thermal expansion coefficients of MnAs (Ref. 5) are about one order of magnitude larger than that of the GaAs substrate.[16] In the course of the α-/β-phase transition, the equilibrium lattice spacing abruptly increases solely along the $[11\bar{2}0]$ direction (a-axis), leading to a net compressive stress in the α-phase.[5] In the coexistence region, the strain is reduced by formation of the periodic α-/β-stripe pattern. In the perpendicular direction (c-axis), on the other hand, no comparable strain relaxation mechanism exists. This also limits the maximum thickness of the epitaxial MnAs films to roughly 500 nm, above which already as-grown films exhibit cracks aligned in the $[11\bar{2}0]$ direction.[17] As the MnAs film expands in the vicinity of the cracks in height by about 4 nm and as the distortion of the α-stripe decays over about 500 nm in the MnAs[0001] direction away from the crack, the strain in the MnAs film is relaxed in both the MnAs[0001] and the $[11\bar{2}0]$ direction. Therefore, the (partial) strain relief owing to the introduction of cracks can be held responsible for the shift of T^* towards the T_c of unstrained bulk MnAs.

In conclusion, we present AFM and MFM investigations around the phase transition temperature of patterned epitaxial MnAs films on GaAs(001). By temperature dependent measurements, we studied the evolution of the MnAs α- and β-phase approaching the transition temperature T^*. Investigations on cracked and uncracked surface regions revealed that T^* is larger in the vicinity of the cracks. According to the strain dependence of the transition temperature and confirmed by the shift of T^* toward the bulk value T_c of MnAs, the strain is locally relieved in these areas. This underlines the important role of the altered strain distribution on the magnetic properties of nanostructures.

We acknowledge C. Herrmann and M. Kästner for sample growth, E. Wiebicke for sample preparation, and M. Ramsteiner for critically reading the manuscript.

[1] D. A. Thompson and J. S. Best, IBM J. Res. Dev. **44**, 311 (2000).
[2] G. Wedler, J. Walz, A. Greuer, and R. Koch, Phys. Rev. B **60**, R11313 (1999).
[3] M. Tanaka, J. P. Harbison, and G. M. Rothberg, J. Cryst. Growth **150**, 1132 (1995).
[4] F. Schippan, A. Trampert, L. Däweritz, and K. H. Ploog, J. Vac. Sci. Technol. B **17**, 1716 (1999).
[5] B. T. M. Willis and H. P. Rooksby, Proc. Phys. Soc. London, Sect. B **67**, 290 (1954).
[6] V. M. Kaganer, B. Jenichen, F. Schippan, W. Braun, L. Däweritz, and K. H. Ploog, Phys. Rev. Lett. **85**, 341 (2000).
[7] V. M. Kaganer, B. Jenichen, F. Schippan, W. Braun, L. Däweritz, and K. H. Ploog, Phys. Rev. B **66**, 045305 (2002).
[8] T. Plake, T. Hesjedal, J. Mohanty, M. Kästner, L. Däweritz, and K. H. Ploog, Appl. Phys. Lett. **82**, 2308 (2003).
[9] A. K. Das, C. Pampuch, A. Ney, T. Hesjedal, L. Däweritz, R. Koch, and K. H. Ploog, Phys. Rev. Lett. **91**, 087203 (2003).
[10] Resist: 130 nm poly(methylmethacrylate), etch solution: $HCl:H_2O_2:H_2O = 4:1:12$
[11] Y. Takagaki, E. Wiebicke, T. Hesjedal, H. Kostial, C. Herrmann, L. Däweritz, and K. H. Ploog, Appl. Phys. Lett. (to be published).
[12] Y. Takagaki, E. Wiebicke, M. Ramsteiner, L. Däweritz, and K. H. Ploog, Appl. Phys. A: Mater. Sci. Process. **76**, 837 (2003).
[13] F. Schippan, G. Behme, L. Däweritz, K. H. Ploog, B. Dennis, K.-U. Neumann, and K. R. A. Ziebeck, J. Appl. Phys. **88**, 2766 (2000).
[14] N. Menyuk, J. A. Kafalas, K. Dwight, and J. B. Goodenough, Phys. Rev. B **177**, 942 (1969).
[15] C. P. Bean and D. S. Rodbell, Phys. Rev. **126**, 104 (1962).
[16] *Handbook of Chemistry and Physics*, 72nd ed., edited by D. R. Lide (CRC, Boston, 1991), pp. 12–68.
[17] L. Däweritz, F. Schippan, A. Trampert, M. Kästner, G. Behme, Z. M. Wang, M. Moreno, P. Schützendübe, and K. H. Ploog, J. Cryst. Growth **227–228**, 834 (2001).