

Strain redistribution at the phase transition of MnAs/GaAs(001) films

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We investigated the thermal evolution of the lattice parameters of a MnAs film epitaxially grown on GaAs(001) around its magnetostructural first-order phase transition using x-ray diffraction. Despite the substrate constraint, large variation of one of the in-plane lattice parameters is preserved, typical of bulk MnAs phase transition, during a large temperature range where two phases coexist. We demonstrated that the condition of the constant film length along this direction, in accord to the substrate length, is always fulfilled during the process. The effect is attributed to the gliding of misfit dislocations present on the film. © 2006 American Institute of Physics. [DOI: 10.1063/1.2194407]

Hybrid semiconductor-ferromagnetic structures have been subject to intensive studies over the last years due to their potential application on spintronics.¹ In particular, MnAs films epitaxially grown on GaAs are strong candidates for spin injection structures² that can be easily integrated to III-V devices.^{3,4} Bulk MnAs presents an abrupt first-order phase transition at $\sim 40^\circ\text{C}$ from a ferromagnetic/hexagonal α phase to a paramagnetic/orthorhombic β phase when the temperature increases. The hexagonal basal plane significantly contracts, while the lattice parameter along its perpendicular direction remains almost unaltered. For thin MnAs films on GaAs(001), this transition loses its abruptness, presenting a coexistence of the two phases as periodic alternated stripes for a large temperature range of at least 20°C .^{5,6} As a result, a fraction of the MnAs film usually remains on the β phase until room temperature, which is undesirable for device applications. Higher phase transition temperatures can be obtained changing the crystal orientation⁷ of the MnAs film on the substrate or with postgrowth annealing.^{8,9} These effects are explained by the change of the film residual strain that strongly affects the MnAs phase transition. The magnetoelastic energy balance governing the phase transition is a rather complex issue that can, however, be approximately expressed by a linear dependence on the MnAs lattice parameters.¹⁰ Therefore, a more detailed understanding of this effect, including an investigation of the actual strains during the phase transition is fundamental for the research on the magnetic properties of MnAs films.

Phase coexistence during a first-order phase transition is explained by the built-in strain created in the MnAs film due to the constraint of the substrate. The main features of this coexistence have been well described by a model based on the minimization of the elastic energy.^{6,11} This theoretical model considers that during the first-order phase transition the strain can be redistributed and only the mean strain over the whole film is constant. During the film growth process at 230°C misfit dislocations are generated due to the large mismatch between the film and the substrate. At room temperature where the first-order phase transition occurs, the

strain redistribution is attributed to the gliding of existing misfit dislocations along the film interface as they cannot be generated at this temperature.^{6,11} We have followed both the out-of-plane and the in-plane lattice parameters of a MnAs film by using x-ray diffraction during the temperature driven phase transition. We observed that, despite the substrate constraint, the in-plane lattice parameter of the film lying on the hexagonal basal plane does vary during phase transition, revealing a rather complex mechanism behind the formation of α and β phase stripes, but keeping a constant mean strain over the whole film.

Our measurements have been performed in a 130 nm thick MnAs film grown on a GaAs(001) substrate at 230°C by molecular beam epitaxy with the A orientation¹² as shown in Fig. 1(a). We use here the orthorhombic crystallographic

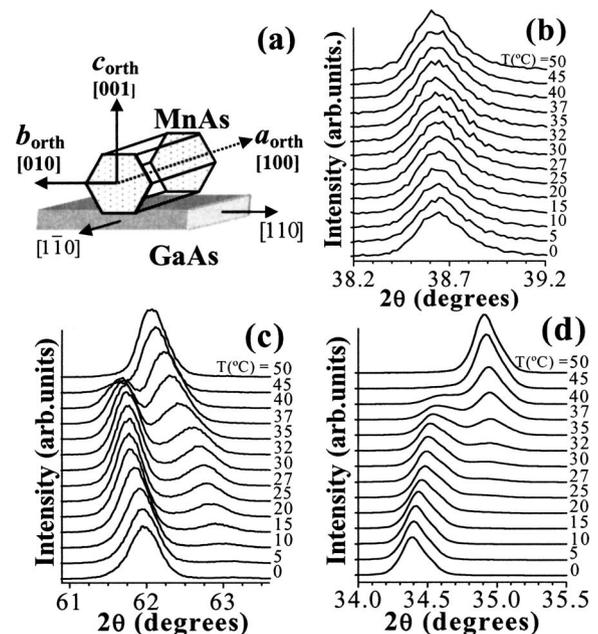


FIG. 1. (a) Scheme of a MnAs film with the A orientation relative to the GaAs(001) substrate. The MnAs lattice parameters (a_{orth} , b_{orth} , and c_{orth}) are defined following the orthorhombic structure notation. Temperature dependent x-ray diffraction patterns obtained during a heating cycle of the sample for reflections (200) (b) and (020) (c) in grazing incidence geometry, and (002) in specular geometry (d).

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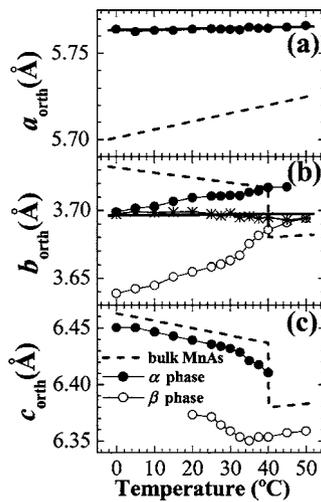


FIG. 2. Lattice parameters: (a) a_{orth} , (b) b_{orth} , and (c) c_{orth} as a function of temperature for α and β phases of the film and bulk MnAs. The solid line in Figs. 2(a) and 2(b) corresponds to a straight line with an angular coefficient given by the GaAs thermal expansion coefficient. The star dots in Fig. 2(b) correspond to the values of the averaged lattice parameter $\langle b_{\text{orth}} \rangle$ presented in the Fig. 3(b).

system for the MnAs film, which is more appropriate since the residual strain breaks off the hexagonal symmetry of the α phase. The out-of-plane and in-plane lattice parameters were, respectively, determined by specular and grazing incidence x-ray diffraction measurements at the Brazilian National Synchrotron Light Laboratory (LNLS) using a monochromatic beam of 6500 eV.¹³ The sample temperature was varied using a closed-cycle He cryostat. The experiments were performed during heating and cooling of the sample, since a thermal hysteresis has been observed in this system.^{7,11} This effect is usually sample (thickness and preparation condition) dependent and in our case, we only observed a minor hysteresis that does not affect our main conclusions. Therefore, we only present the results for heating cycles.

Figures 1(b)–1(d) show the x-ray diffraction spectra obtained at different temperatures during a heating cycle, for different reflections. Both the (020) and the (002) reflections [Figs. 1(c) and 1(d)] show two distinct peaks attributed to the different lattice parameters of the α and β phases, which coexist for a large temperature range. The observation of two distinct out-of-plane lattice parameters for the unconstrained [001] direction is fully expected and has been intensively investigated before,^{6,7,11} but it is rather surprising for the in-plane lattice parameters in the [010] direction, which is constrained by the GaAs substrate. For the (200) reflection [Fig. 1(b)], we only observe a single peak, which is in agreement with the negligible change of the corresponding lattice parameter for bulk MnAs.

The temperature evolution of the lattice parameters: a_{orth} , b_{orth} , and c_{orth} , obtained from our measurements are presented in Fig. 2. We also present the lattice parameter of bulk MnAs (dashed lines) and a solid straight line with an angular coefficient corresponding to the thermal expansion coefficient of the GaAs substrate, for the in-plane directions [Figs. 2(a) and 2(b)]. The MnAs unit cell is markedly elongated along the a_{orth} -axis relative to the bulk ($\sim 1\%$ strain). As mentioned above, a_{orth} assumes a single value for both phases, varying only weakly at the analyzed temperature

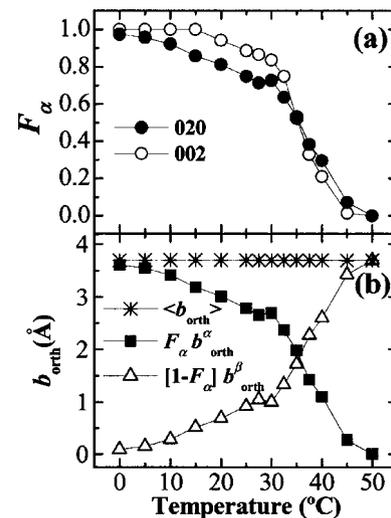


FIG. 3. (a) Fraction of the α MnAs phase as a function of temperature obtained from the (020) and (002) reflections; (b) Temperature dependence of parameters b_{orth}^{α} and b_{orth}^{β} weighted by the corresponding fraction of each phase and the resulting averaged value of b_{orth} for the film.

range and roughly following the thermal expansion of the GaAs (solid line), as expected for an in-plane lattice parameter constraint by the substrate. On the other hand, the out-of-plane lattice parameter c_{orth} [in Fig. 2(c)] is slightly compressed as compared to the bulk. Otherwise, the general behavior of c_{orth} for the film is quite similar to the bulk, as expected for this unrestrained direction. For low temperatures when only the α phase is present, c_{orth} shows a thermal contraction with a coefficient similar to that of bulk α MnAs, while it expands with a coefficient similar to bulk β MnAs for temperatures larger than 40 °C, when only the β phase is present. During phase coexistence, two distinct lattice parameters corresponding to α and β phases are observed with a relative difference similar to that observed for the bulk phase transition at 40 °C.

Even though b_{orth} , as well as a_{orth} , represents an in-plane lattice direction and should therefore be subjected to the GaAs substrate constraint, we observe two distinct values of b_{orth} , corresponding to α and β phases, and their relative difference is also similar to that observed for the bulk MnAs transition at 40 °C. Furthermore, b_{orth} presents a large and unusual variation as a function of temperature, for both phases, including some persistence of the α phase above 40 °C and of the β phase below 20 °C. The total variation of this lattice parameter when the film changes from a fully α phase in 0 °C to a fully β phase condition in 55 °C [$b_{\text{orth}}(T=0\text{ °C}) - b_{\text{orth}}(T=55\text{ °C})$], approximately corresponds to the GaAs thermal expansion for this temperature interval (solid line). This result demonstrates that the film maintains the lateral dimension of the GaAs substrate beyond the edges of the phase transition. In order to analyze if this condition is also fulfilled during the phase transition, we have to consider the fraction of volume of each phase at a given temperature. Neglecting small differences of structure factors for equivalent reflections for each phase, these fractions are proportional to the integrated intensity of the corresponding x-ray peaks. We thus obtained the fraction F_{α} of the α phase versus temperature from the (020) and (002) reflections, which are presented in Fig. 3(a). We observe a small discrepancy between F_{α} obtained for those two reflections

tions whose origin is not completely understood but could be originated from the structure factor correction, which are difficult to determine for these strained structures.

It has been shown^{5,14} that the α and β phase stripes formed during the phase coexistence on the film are extended along the [100] direction and periodic along the [010] direction. Considering also that the α and β volumes are continuous along the film thickness (growth direction), i.e., for a given α (β) cell, all the cells along the [100] and [001] directions passing through this given cell are also at the α (β) phase. In this case, the total length $l_{[010]}(T)$ of the film along the [010] direction should thus be proportional to the averaged lattice parameter along this direction considering the fractions of each phase: $l_{[010]}(T) = N_{[010]}(b_{\text{orth}}^{\alpha}(T)F_{\alpha}(T) + b_{\text{orth}}^{\beta}(T)[1 - F_{\alpha}(T)])$, where $N_{[010]}$ corresponds to the number of unit cells of the film along the [010] direction. In Fig. 3(b) we present the averaged lattice parameter $\langle b_{\text{orth}} \rangle = (l_{[010]}/N_{[010]})$, and also the weighted lattice parameters for each phase, $F_{\alpha}(T)b_{\text{orth}}^{\alpha}(T)$ and $b_{\text{orth}}^{\beta}(T)[1 - F_{\alpha}(T)]$. The values obtained for $\langle b_{\text{orth}} \rangle$ are also shown in Fig. 2(b) in a more detailed scale, which clearly demonstrates that despite the strong variation of b_{orth} with temperature, the total length along this direction does remain constant during phase transition.

We point out that we can also estimate from our results the temperature where the density of misfit dislocations in the film freezes out. At the growth temperature, it is reasonable to assume that the lattice mismatch between MnAs and GaAs is completely relaxed by misfit dislocations.¹⁵ As the sample is cooled down, strain should start to build up due to the distinct thermal coefficients of those materials, starting at a critical temperature where the thermal energy becomes insufficient to change the density of dislocations and thus relieve the strain. Below that temperature, the in-plane parameters of the MnAs film must follow the corresponding GaAs thermal expansion coefficient. This critical temperature can thus be estimated by the crossing point of the MnAs and GaAs thermal expansions represented, respectively, by the dashed and the continuous lines in Figs. 2(a) and 2(b). We obtained a critical temperature of ~ 150 °C for both cases.¹⁵ We remark that during the first-order phase transition the density of dislocations should thus remain constant, but those dislocations must be allowed to glide to provide the degree of freedom that results on the observed lattice parameter

variation of the MnAs film along the [010] direction.

In summary, we have performed a detailed study of the temperature dependence of the lattice parameters of a MnAs/GaAs film during phase transition. Our measurements revealed an unexpected large variation of one of the in-plane lattice parameters during α and β phase coexistence. The film maintains, however, a constant total length along this direction during all phase transition, which is consistent with the partial constrain considered by theoretical models and justified by the gliding of misfit dislocations present on the film.

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- ¹I. Žutić, J. Fabian, and S. Das Sarma, *Rev. Mod. Phys.* **76**, 323 (2004).
- ²M. Ramsteiner, H. Y. Hao, A. Kawaharazuka, H. J. Zhu, M. Kästner, R. Hey, L. Däweritz, H. T. Grahn, and K. H. Ploog, *Phys. Rev. B* **66**, 081304 (2002).
- ³M. Tanaka, J. P. Harbinson, T. Sands, T. L. Cheeks, V. G. Keramidias, and G. M. Rothberg, *J. Vac. Sci. Technol. B* **12**, 1091 (1994).
- ⁴M. Tanaka and J. P. Harbinson, *Appl. Phys. Lett.* **65**, 1964 (1994).
- ⁵T. Plake, M. Ramsteiner, V. M. Kaganer, B. Jenichen, M. Kästner, L. Däweritz, and K. H. Ploog, *Appl. Phys. Lett.* **80**, 2523 (2002).
- ⁶V. M. Kaganer, B. Jenichen, F. Schippan, W. Braun, L. Däweritz, and K. H. Ploog, *Phys. Rev. Lett.* **85**, 341 (2000).
- ⁷F. Iikawa, M. J. S. P. Brasil, O. D. D. Couto, C. Adriano, C. Giles, and L. Däweritz, *Appl. Phys. Lett.* **85**, 2250 (2004).
- ⁸J. H. Song, Y. Cui, J. J. Lee, and J. B. Ketterson, *Appl. Phys. Lett.* **87**, 092504 (2005).
- ⁹L. Däweritz, C. Herrmann, J. Mohanty, T. Hesjedal, K. H. Ploog, E. Bauer, A. Locatelli, S. Cherifi, R. Belkhou, A. Pavlovska, and S. Heun, *J. Vac. Sci. Technol. B* **23**, 1759 (2005).
- ¹⁰F. Iikawa, M. J. S. P. Brasil, C. Adriano, O. D. D. Couto, C. Giles, P. V. Santos, L. Däweritz, I. Rungger, and S. Sanvito, *Phys. Rev. Lett.* **95**, 077203 (2005).
- ¹¹V. M. Kaganer, B. Jenichen, F. Schippan, W. Braun, L. Däweritz, and K. H. Ploog, *Phys. Rev. B* **66**, 045305 (2002).
- ¹²F. Schippan, A. Trampert, L. Däweritz, and K. H. Ploog, *J. Vac. Sci. Technol. B* **17**, 1716 (1999).
- ¹³C. Giles, F. Yokaichia, S. W. Kycia, L. C. Sampaio, D. C. Ardiles-Saraiva, M. K. K. Franco, and R. T. Neuenschwander, *J. Synchrotron Radiat.* **10**, 430 (2003).
- ¹⁴M. Kästner, C. Herrmann, L. Däweritz, and K. H. Ploog, *J. Appl. Phys.* **92**, 5711 (2002).
- ¹⁵F. Iikawa, P. V. Santos, M. Kästner, F. Schippan, and L. Däweritz, *Phys. Rev. B* **65**, 205328 (2002).