

Stress evolution during Fe(001) epitaxy on GaAs(001)

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We report on *in situ* stress measurements of Fe/GaAs(001), which enlighten the dynamics of the interface formation of this important magnetic metal/semiconductor system. At deposition temperatures of 300 and 450 K, the stress evolution during growth is very similar. In Fe films thicker than 6–7 nm, the stress is compressive owing to the misfit between the lattices of Fe and GaAs. Thinner films surprisingly are dominated by a tensile stress contribution due to considerable As (and Ga) interdiffusion even at 300 K. © 2001 American Institute of Physics.
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Thin Fe films on GaAs substrates have evolved as a model system for the integration of magnetic materials with semiconductors. On GaAs(001) epitaxial body-centered-cubic (bcc) Fe(001) films are obtained with Fe[001] being parallel to GaAs[001].¹ In the majority of investigations Fe has been deposited at elevated temperatures (420–450 K). However, due to the diffusion of Ga and As into the Fe film² no sharp interface is formed with detrimental consequences for the magnetic moment of the first few nanometers.^{2,3} Irrespective of the actual substrate configuration — Ga rich (4×6)³ or As rich (2×4) and $c(4\times 4)$ ^{2,4} — a thin layer of $\text{Fe}_3\text{Ga}_{2-x}\text{As}_x$ has been detected at the metal/semiconductor interface exhibiting only half the magnetization of bulk Fe.⁵ Arsenic also diffuses into thicker film regions and segregates to the surface.²

In order to reduce intermixing effects Fe was also recently deposited at lower temperatures. On the Ga rich (4×6) surface good epitaxial growth has been achieved at room temperature.^{6–8} The onset of magnetization was found to occur at about 3 monolayers (ML) after passing an initial three dimensional island stage.^{7,8} Contrary to the high temperature studies the films recover full magnetic moment without formation of a dead magnetic layer.^{6,7} Whereas As and Ga diffusion is observed also in the room temperature experiments,^{9,8} no $\text{Fe}_3\text{Ga}_{2-x}\text{As}_x$ compound has been detected at the interface.^{10,11}

Here we report on real time stress measurements of Fe/GaAs(001) at 300 and 450 K, which enlighten the dynamics of the interface formation. At both temperatures the evolution of stress with film thickness is very similar. In Fe films thicker than 6–7 nm the stress is compressive owing to the misfit between the lattices of Fe ($a=0.2866$ nm) and GaAs ($a/2=0.2827$ nm). Thinner films are surprisingly dominated by a tensile stress contribution, which points to considerable intermixing at the interface even at 300 K in contrast to the previous findings.

The experiments were performed in a multiple chamber ultrahigh vacuum (UHV) system (base pressure $<10^{-10}$ mbar) equipped with a sensitive cantilever beam magneto-

meter (CBM) for *in situ* stress and magnetic measurements,¹² a four-grid low energy electron diffraction (LEED) optics for *in situ* control of the substrate and film quality as well as a home-built UHV scanning tunneling microscope (STM) for *in situ* structural investigations. The substrates were cut from ≈ 100 μm thick As capped GaAs(001) wafers, which were prepared in a separate molecular beam epitaxy (MBE) chamber by removal of the oxide, deposition of an ≈ 0.5 μm thick GaAs(001) buffer layer and coating with a thin As layer. Unless stated otherwise, the substrates were further prepared — similar to previous studies^{7,13} — by desorbing the As capping layer around 650 K and annealing for 1 h at 810 K prior to deposition. Fe was electron beam evaporated from a Knudsen-type tungsten source at a pressure better than 2×10^{-9} mbar; the deposition rate determined by a quartz crystal microbalance was 0.008 ± 0.001 nm/s. In each experimental run a CBM and an STM substrate were prepared simultaneously, i.e., under identical conditions. Immediately after the film preparation the STM sample was transferred to the STM chamber without breaking UHV and imaged at room temperature. The magnetic measurements were performed *in situ* with the CBM sample.

The structural and magnetic properties of our Fe films are fully consistent with the previous studies. At both 300 and 450 K the films exhibit good quality LEED patterns of Fe(001) [e.g., Fig. 1(a)]. STM reveals a mounded surface morphology [e.g., Fig. 1(b)] with an average surface roughness of about 1 nm in agreement with a previous STM study.⁴ At 25 nm the film magnetization is 1.9 MA/m and 1.7

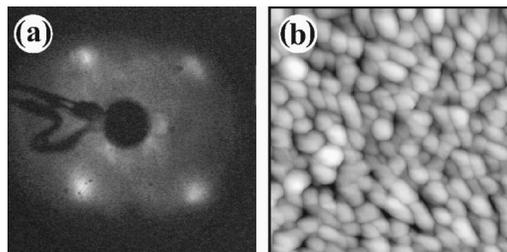


FIG. 1. Fe/GaAs(001) at 300 K: (a) LEED pattern at a primary energy of 174 eV and (b) 80×80 nm² STM topview of a 25 nm thick Fe film.

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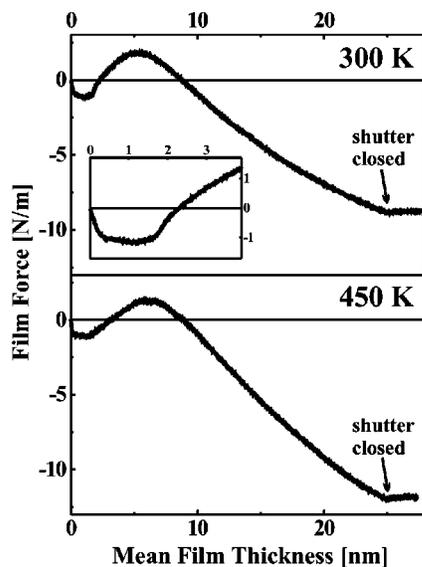


FIG. 2. Film forces (i.e., integral forces in films of unit width) measured in real time during the deposition of Fe onto GaAs(001) at 300 and 450 K.

MA/m at 300 and 450 K, respectively. The absolute values of both films therefore are bulklike (1.76 MA/m) within experimental error ($\approx 10\%$); the magnetization of the 450 K films, however, is always tendentially smaller.

Figure 2 shows the film forces (i.e., the integral forces in films of unit width) as a function of the average film thickness, measured in real time during the Fe deposition at 300 and 450 K. Both films exhibit a nearly identical stress behavior which is characterized by the following features: (i) large compressive forces of about 1.5 N/m at the very beginning of growth, i.e. while depositing the first 2–3 ML of Fe, (ii) a tensile stress contribution in the thickness range of 2–6 nm, and (iii) eventually compressive stress of about 1 GPa in the thicker films.

Our experiments indeed reveal a rather complex stress behavior of Fe/GaAs(001), particularly at film thicknesses below 6 nm. The stress in the thick films originates from the misfit between Fe(001) and GaAs(001). In the case of ideal coherent growth the misfit of 1.36% would lead to compressive stress of -2.8 GPa. The instantaneous stress of 0.7 and 0.9 GPa determined from the slope after the maximum of the force curves at 300 and 450 K, respectively, are significantly smaller than the ideal misfit stress indicating partial strain relief by misfit dislocations. The residual strain of 0.4%–0.45% calculated from the stress values is in reasonable agreement with the 0.6% reported recently from Fe films deposited *in situ* onto MBE-prepared substrates.¹¹ Note that in thicker films the slope of the force curves gradually decreases as compressive strain is further relieved by incorporation of defects. Due to the increased mobility film growth is more perfect at 450 K, thus promoting the transmittance of the compressive strain field.

Compressive stress is also observed at the very beginning of the Fe deposition. The film forces developing in the first 2–3 ML correspond to compressive stress of -3.5 GPa, i.e., significantly larger than the value of the maximum possible misfit stress. Since there is no abrupt transition from the GaAs to the Fe lattice, a plain application of misfit considerations is not appropriate in the case of very thin Fe films on

GaAs(001). STM investigations of GaAs(001) (2×4) revealed that nucleation of Fe proceeds preferentially by breaking As dimer bonds and forming extended Fe–As–Fe chains.⁴ A similarly mixed interlayer phase certainly is formed also on the Ga-rich substrates. Therefore the stress evolving initially is better understood in terms of surface stress changes occurring when the substrate reconstruction is disassembled and the new interface, composed of Fe, Ga, and As atoms, is formed. Surface stress changes of the order of a few N/m upon adsorption indeed are quite common¹⁴ and have been reported previously, e.g., for Ga/Si(001)¹⁵ or Fe/W(110).¹⁶

Subsequent to the nucleation stage, at 2–6 nm, the overall stress is dominated by a tensile component. Tensile stress in the Fe/GaAs(001) system is indeed surprising. Its appearance coincides with the thickness range, where — according to the literature^{2,5} — As and, though to a lesser extent, Ga are predominantly diffusing into the Fe film. The tensile stress contribution therefore seems to be directly related to these diffusion processes. From its sign — tensile and not compressive — the location of the stressed region can be inferred. For instance, As atoms occupying interstitial sites of the Fe lattice — as speculated previously⁹ — would give rise to compressive stress. Also, substitution of bcc Fe atoms by As yields compressive stress, because the volume of possible Fe–As species, such as Fe₂As, FeAs, or FeAs₂, is larger by about 4% than that of pure Fe units.¹⁷ Tensile stress, on the other hand, is generated when As or Ga atoms diffuse out of the GaAs matrix and leave voids behind. A tensile stress component remains, even when the voids are refilled by Fe, because of the smaller volume of Fe compared with As or Ga. In both cases the attractive forces obviously overcompensate the compressive stress contributions of the As and Ga atoms in the Fe film. Notice that further diffusion steps of As or Ga within the Fe lattice do not change the total stress as the stressed sites are moving with the diffusing atoms. Our experiments therefore provide strong evidence that the tensile stress region is confined to a thin layer at the interface.

In order to get a quantitative estimate of the tensile interface stress, we make use of the x-ray absorption fine-structure spectroscopy results of Gordon *et al.*¹⁰ They found that also the Fe lattice of a 10 ML (≈ 1.5 nm) thick films prepared at room temperature is compressed, namely by 1.1%, thus suggesting that the total stress measured in the thickness range of 2–6 nm is the sum of two components: compressive misfit stress and tensile interface stress. Whereas the source for the tensile stress depletes at about 6 nm, the Fe lattice also remains compressed in thicker films (compare Ref. 11). For our estimation we assume that the Fe lattice dimensions of thicker films also hold for the thinner ones, i.e., that the same compressive stress is also experienced by thinner films. Extrapolating the compressive force of the thick films (with the slope after the force curve maximum) back to the zero thickness therefore yields the total tensile force component (see Fig. 3). It is 9 N/m for the 450 K film and 6.5 N/m for the 300 K film implying that the diffusion processes are not inhibited at 300 K. Though being reduced, they still play an important role in the interface formation.

In previous studies ordered phases of Fe₂Ga₂As₂ and

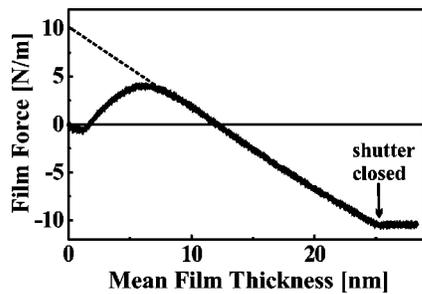


FIG. 3. Film forces (i.e., integral forces in films of unit width) measured in real time during the deposition of Fe onto As-rich GaAs(001) at 300 K; dashed line is used to estimate tensile interface stress.

FeAs₂ have been detected in Fe films prepared at 450 K.¹¹ Particularly the former compound was found to be mainly located at the metal/semiconductor interface,⁵ in agreement with high resolution x-ray photoelectron spectroscopy which shows a strong attenuation of the Ga signal with Fe thickness.^{2,9} In view of these findings we conclude that these ordered phases are responsible for the tensile interface stress observed at 450 K. At lower temperatures the interface is still composed of a mixture of Fe, As, and Ga atoms, however, mainly distributed randomly at 300 K so that no diffraction signal is obtained.¹¹ We remark that tensile stress may also arise from the As atoms that have been shown to segregate to the surface regardless of the substrate reconstruction or preparation.^{2,8,9} In the case of As/Si(001) tensile surface stress of 1.4 N/m was found for a full As monolayer.¹⁸ On Fe/GaAs(001) the expected tensile surface stress therefore is at least one order of magnitude smaller than the observed stress contribution. By assuming a reasonable average tensile strain of only 2% in the interface region the thickness of the involved intermixed layer is estimated to 3–4 nm.

In order to test our interpretation that the observed tensile stress is mainly related to As out diffusion, we prepared “As-richer” substrates by desorbing the As capping layer until the appearance of (001) LEED spots, but omitting the subsequent annealing step. These substrates are terminated mainly by As dimers,¹⁹ rather than by Ga dimers as in Ga rich surfaces.²⁰ Figure 3 shows the force curve measured during the subsequent room temperature deposition of an epitaxial Fe(001) film. The force curve still exhibits the same features as found on the annealed substrates (Fig. 2) with compressive stress of 0.85 GPa in the thick film. The tensile force component, however, has increased to about 10 N/m (compared to 6.5 N/m on the annealed surface) thus confirming the key role of As diffusion. It is noteworthy that the film forces are constant immediately after the Fe shutter is closed

(see Figs. 2 and 3). Obviously the amount of As (and Ga) diffusing into the Fe film is limited and exhausted at a thickness of about 6 nm.

In conclusion, our real time stress measurements of Fe/GaAs(001) surprisingly revealed a tensile stress contribution which exceeds the compressive misfit stress in thin films. As inferred from its sign, this stress contribution is related to the restructuring of the interface proceeding when (preferentially) As diffuses into the growing Fe film. Compared to the 450 K film the tensile stress contribution is only reduced by about 30% at 300 K, thus demonstrating that the diffusion induced restructuring of the interface is not frozen out at 300 K. As diffraction experiments are sensitive mainly to crystallographically ordered structures stress measurements provide important complementary information on the temperature dependent growth of Fe/GaAs(001).

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- ¹J. J. Krebs, B. T. Jonker, and G. A. Prinz, *J. Appl. Phys.* **61**, 2596 (1987).
- ²E. M. Kneedler, B. T. Jonker, P. M. Thibado, R. J. Wagner, B. V. Shanabrook, and L. J. Whitman, *Phys. Rev. B* **56**, 8163 (1997).
- ³M. Gester, C. Daboo, R. J. Hicken, S. J. Gray, A. Ercole, and J. A. C. Bland, *J. Appl. Phys.* **80**, 347 (1996).
- ⁴P. M. Thibado, E. Kneedler, B. T. Jonker, B. R. Bennet, B. V. Shanabrook, and L. J. Whitman, *Phys. Rev. B* **53**, R10481 (1996).
- ⁵A. Filipe, A. Schuhl, and P. Galtier, *Appl. Phys. Lett.* **70**, 129 (1997).
- ⁶M. Zöphl, M. Brockmann, M. Köhler, S. Kreuzer, T. Schweinböck, S. Miethaner, F. Bensch, and G. Bayreuther, *J. Magn. Magn. Mater.* **175**, 16 (1997).
- ⁷Y. B. Xu, E. T. M. Kernohan, D. J. Freeland, A. Ercole, M. Tselepi, and J. A. C. Bland, *Phys. Rev. B* **58**, 890 (1998).
- ⁸T. L. Monchesky, B. Heinrich, R. Urban, K. Myrtle, M. Klaua, and J. Kirschner, *Phys. Rev. B* **60**, 10 242 (1999).
- ⁹S. A. Chambers, F. Xu, H. W. Chen, I. M. Vitomirov, S. B. Anderson, and J. H. Weaver, *Phys. Rev. B* **34**, 6605 (1986).
- ¹⁰R. A. Gordon, E. D. Crozier, D.-T. Jiang, T. L. Monchesky, and B. Heinrich, *Phys. Rev. B* **62**, 2151 (2000).
- ¹¹H.-P. Schönherr, R. Nötzel, W. Ma, and K. H. Ploog, *J. Appl. Phys.* **89**, 169 (2001).
- ¹²M. Weber, R. Koch, and K. H. Rieder, *Phys. Rev. Lett.* **73**, 1166 (1994).
- ¹³D. E. Bürgeler, C. M. Schmidt, J. A. Wolf, T. M. Schaub, and H.-J. Güntherodt, *Surf. Sci.* **366**, 295 (1996).
- ¹⁴H. Ibach, *Surf. Sci. Rep.* **29**, 193 (1997).
- ¹⁵R. E. Martinez, W. M. Augustyniak, and J. A. Golovchenko, *Phys. Rev. Lett.* **64**, 1035 (1990).
- ¹⁶D. Sander, R. Skomski, C. Schmidthal, A. Enders, and J. Kirschner, *Phys. Rev. Lett.* **77**, 2566 (1996).
- ¹⁷R. W. G. Wyckoff, *Crystal Structures*, 2nd ed. (Wiley Interscience, New York, 1963), Vol. 1.
- ¹⁸A. J. Schell-Sorokin and R. M. Tromp, *Surf. Sci.* **319**, 110 (1994).
- ¹⁹V. P. LaBella, H. Yang, D. W. Bullock, P. M. Thibado, P. Kratzer, and M. Scheffler, *Phys. Rev. Lett.* **83**, 2989 (1999).
- ²⁰S.-H. Lee, W. Moritz, and M. Scheffler, *Phys. Rev. Lett.* **85**, 3890 (2000).