

Growth and Magnetic Properties of Epitaxial Fe(100) on S-Passivated GaAs(100)

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This paper proposes a new technique for the epitaxial growth of Fe on GaAs(100) which eliminates the problems of substrate interdiffusion through the overlayer. S-passivation of the GaAs(100) surface prior to Fe overgrowth is shown to be an effective way of inhibiting the interdiffusion of As and Ga into the Fe overlayer. The resulting Fe layer is found to grow in a bcc (100) orientation and is ferromagnetic for coverages >4 monolayers.

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The growth of magnetic overlayers and multilayers on semiconductors has attracted considerable attention due to the potential for integrating the desired properties of magnetic thin film systems with computer circuitry. In particular, magnetic multilayer systems composed of alternating layers of magnetic and nonmagnetic materials can exhibit magnetic properties which differ significantly from those of the corresponding bulk materials due to interfacial effects and low dimensionality [1]. One such property is the giant magnetoresistance (GMR) effect, whereby the electrical resistance of a sample can be significantly changed by the application of a magnetic field. This is of practical interest due to applications in high density magnetic storage devices and magnetic sensors.

However, in practice, the growth of transition metals on semiconducting substrates is problematic, generally resulting in the intermixing of the semiconducting elements with the overlayer and often resulting in disordered overlayers. The possibility of growing Fe overlayers on GaAs is particularly promising due to the low lattice mismatch (1.3% with 2 Fe atoms per substrate surface atom) of the system and the observation that antiferromagnetically coupled Fe layers can exhibit the giant magnetoresistance effect. Previous studies on this system indicate that bcc Fe grows epitaxially on GaAs(100) for thicknesses up to 300 Å. However, the Fe film is not flat, and As is intermixed with the Fe film as well segregated on the surface [2–4]. This As interdiffusion is believed to be the cause of a dramatic reduction of the magnetization of the Fe overlayer on the GaAs(100) surface [4].

The growth of an Ag buffer layer on the GaAs substrate prior to the Fe deposition has been utilized to stop the As interdiffusion [5]. The use of an Ag buffer layer introduces difficulties in that the buffer layer (typically >100 Å) induces the tilted growth of the Fe overlayer as well as acting as an electrical shunt. The presence of this shunt requires that the magnetic structure be impedance matched to the buffer layer. This would require the growth of thick magnetic superlattices, which presents further problems, as it is difficult to maintain a high

quality of growth for thick samples at the relatively low temperatures required to avoid interface interdiffusion between the magnetic and nonmagnetic layers.

In this paper we propose an alternative solution, examining the growth of Fe overlayers on a S-passivated GaAs(100) surface. To our knowledge this is the first example of the utilization of surface passivation of the semiconductor substrate as a means of inhibiting the diffusion of semiconductor material into a transition metal overlayer. Surface passivation of semiconductors has been well established as a way of improving the electrical properties of devices [6,7]. Various semiconducting films have been grown on passivated semiconductor surfaces [8–10], mostly in attempts to create quantum confinement systems and lasers. Aluminum has also been grown on S-passivated GaAs(100), and it has been suggested that the S hinders chemical reaction between Al and the substrate [11]. However, to date there have not been any attempts to grow transition metals epitaxially on such a passivated substrate.

In this investigation we examined the growth of Fe on S-passivated GaAs(100) with Auger electron spectroscopy (AES), angle resolved AES (ARAES), low energy electron diffraction (LEED), static secondary ion mass spectrometry (SSIMS), and surface magneto-optic Kerr effect (SMOKE) measurements. We will show that bcc Fe(100) grows epitaxially on this substrate, with part of the S layer floating out as an ordered overlayer on top of the Fe film. Diffusion or segregation of As is not observed. The Fe film is shown to be ferromagnetic for Fe coverages as low as 4 monolayers (ML). Possible mechanisms for the inhibition of As diffusion by the S overlayer are also discussed.

The experiments were carried out in a diffusion pumped UHV chamber (the base pressure is less than 1×10^{-10} torr) which is connected to a turbo-pumped UHV load-lock chamber (base pressure 2×10^{-10} torr) equipped with a remote sample transfer arm. The chamber is interfaced, via a two stage differential pumping station, to a 2.5 MV Van de Graaff accelerator which can be utilized for ion beam measurements. The main

growth and analysis chamber is equipped with an Fe evaporation source, a four-grid retarding field analyzer for LEED measurements, a CLAM electron energy analyzer (Fisons Instruments) for AES and ARAES measurements, and a SSIMS system consisting of a Hiden quadrupole mass spectrometer and a Kimball Physics Cs^+ ion gun. The LEED images were collected with a charge coupled device camera from the LEED screen and recorded on video tape. The videocassette recorder is interfaced to a computer system, allowing subsequent analysis of the LEED spot intensities and widths. The load-lock chamber is equipped with a SMOKE apparatus for *in situ* magnetic measurements.

The substrates utilized in this investigation were cut from *n*-type GaAs(100) single crystal wafers from Bertram laboratories. The substrates were prepared by first rinsing in hot (~ 320 K) methanol, and then treating for 20 min in an aqueous $(\text{NH}_4)_2\text{S}$ solution at 340 K. After this treatment the samples were rinsed with running deionized water and static methanol, blown dry with He, and introduced into the load-lock system. Samples prepared in this manner are known to have 1 ML of bridge bonded S on the surface [12,13], and are remarkably stable against oxidation by air. A new substrate was utilized for each experiment.

The as-prepared wafers exhibited a poor (1×1) LEED pattern, and AES measurements indicated the presence of S, Ga, As, and trace amounts of C. After annealing to 720 K for ~ 20 min, a sharp (2×1) LEED pattern (the reconstruction is due to the dimerization of adjacent S rows [14–16]) was observed and the C contamination was removed. The AES S level was monitored during annealing and it indicated that there was no loss of S during the anneal, in agreement with the observations of a previous study [17]. The Fe was deposited from a homemade liquid nitrogen shrouded sublimation source onto the substrate held at room temperature, at a rate of 1.6 ML/min [where 1 ML corresponds to the Fe(100) surface density]. The Fe coverages were determined via a Rutherford backscattering calibration of the evaporation rate onto an Al substrate [18].

The LEED pattern was monitored as Fe was deposited, and the spot intensities for two different spots at 117 eV are presented in Fig. 1. Initially, the S/GaAs(100)- (2×1) pattern was observed to fade while the diffuse background increased significantly, indicating the presence of a disordered interface. By about 3 ML of Fe deposition in a LEED pattern from the Fe overlayer was first observed. The pattern was aligned in the expected manner with the principal crystallographic directions of the overlayer being identical to those of the substrate, indicating that the bcc Fe(100) grows epitaxially on the S/GaAs(100) substrate. The spot intensities were then observed to increase rapidly as further Fe was deposited, and they began to level off at an Fe coverage of about 7 ML. Visual examination of

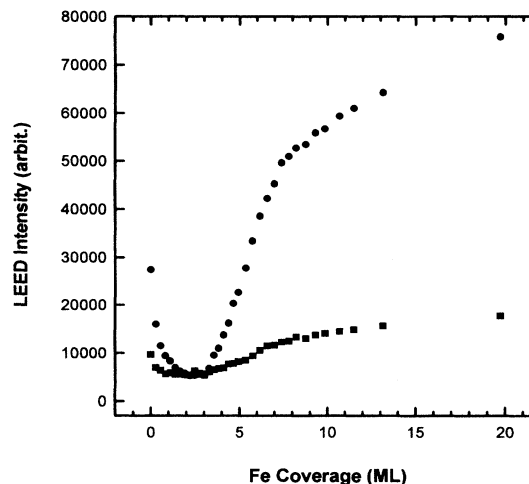


FIG. 1. The spot intensities for two LEED spots during Fe deposition. The circles represent the $(1,1)$ GaAs and $(1,0)$ Fe spots, while the squares represent the $(1,0)$ GaAs and $(\frac{1}{2}, \frac{1}{2})$ S/Fe spots.

the spot intensity maxima as a function of beam energy showed good agreement with the $I(V)$ curves of $c(2 \times 2)$ S/Fe(100) [19] (and are significantly different from those for clean Fe(100) [20]), suggesting that part of the S passivation layer is floating out as an ordered overlayer. S segregation has been confirmed by AES measurements as will be discussed below.

The LEED spots from the Fe overlayer initially were broad, but sharpened to a minimum width between 6 and 12 ML coverage. Further deposition of Fe resulted in a slight broadening of the spots, indicating a reduction in the quality of epitaxy. The widths of the Fe overlayer spots are comparable to those of the substrate before Fe deposition for energies < 120 eV. At higher energies, however, the overlayer spots were significantly broader than those of the substrate, suggesting that the long-range order is not uniform through the entire thickness of the film. The half-order spots, which are due to the S overlayer, were significantly less intense and broader than those from the Fe, indicating that the S overlayer is less well ordered.

ARAES, which has proven to be a powerful tool for characterizing the local structure of overlayer systems [21], was utilized to examine the Fe overlayer. The ARAES scans of the Fe overlayer [18] show that the Fe is growing on a bcc (100) structure on S/GaAs(100). The first forward focusing peaks are observed between 1 and 1.5 ML Fe coverage, with all of the expected maxima for a bcc lattice being observed by 2 ML. This indicates that the growth of Fe is proceeding by a three-dimensional mechanism, at least in the initial growth region. In addition, while the interfacial region may be disordered in terms of long-range order, there is still order in the short-range bonding geometry.

Figure 2 presents the intensity of the Fe, S, Ga, and As AES transitions as a function of Fe coverage. A takeoff angle of 30° was utilized to enhance the surface sensitivity and minimize the forward scattering effects. The Fe signal is observed to increase steadily, reaching saturation at ~ 8 ML coverage. The Ga and As signals decrease exponentially, completely disappearing for Fe coverages higher than 15 ML. Fitting these results to exponentials yielded mean free paths for Ga and As AES electrons of 14 and 19 Å, respectively. These values are in good agreement with the calculated values of 17 and 19 Å [22], suggesting that, after the initial interfacial growth, the Fe film is uniform and neither As nor Ga is interdiffusing through the film. The S signal decreases rapidly for coverages up to 4 ML and then remains relatively constant, at approximately 70% of its initial intensity, upon further Fe deposition. The persistence of a such strong S signal at high Fe coverages despite the low mean free path of these electrons (~ 7 Å) suggests that about 70% of the S floats on top of the film, in agreement with the LEED results. As the S-Fe bond is known to be very strong, this S segregation may help to stabilize the high energy Fe(100) surface.

According to the AES experiments, As does not appear to interdiffuse or segregate through the Fe overlayer. However, due to the low sensitivity for As and the large mean free path of As electrons, AES is not the best technique for determining the presence of As in the overlayer. In order to improve the As sensitivity, SSIMS measurements were performed on Fe films of 2, 4, and 6 ML thicknesses. In each measurement the As SIMS signal was at background levels, with the S and Fe signals being clearly observed. In comparison to the signal levels observed on the clean S/GaAs(100) substrate, we conclude that the upper limit for the amount of As in or on the overlayer is on the order of 0.01 ML.

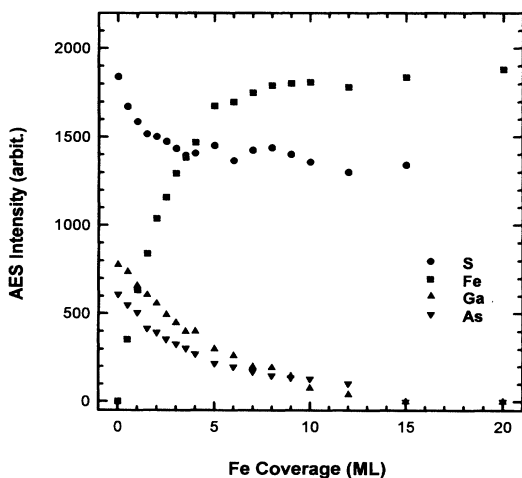


FIG. 2. The S, Fe, Ga, and As AES signals for different coverages of Fe, measured at 30° take-off angle.

Two mechanisms for the inhibition of As diffusion are possible which are consistent with the behavior of the sulphur during Fe deposition. It is possible that the S which remains at the Fe/GaAs interface is preferentially located at defects or other active sites for As diffusion, and thus inhibits As diffusion by blocking the sites. Another possibility is that the S prefers to bond to the Fe overlayer and acts as a surfactant, holding the Fe on top of the substrate. As the Fe layer grows, the S is no longer attached to the Fe at the Fe/GaAs interface. However, As can now only move by bulk diffusion, which will be a highly activated process and may not be energetically feasible at room temperature. Further experiments, which will be reported elsewhere, were performed where elevated temperatures were utilized to ensure that all of the S floated out on top of the Fe overlayer. These experiments strongly suggest that it is this second mechanism (S acting as a surfactant) which is responsible for the prevention of As intermixing.

The magnetic properties of the Fe overlayers were investigated by SMOKE measurements in the longitudinal configuration [23]. Figure 3 displays hysteresis curves measured at room temperature for various Fe coverages with the magnetic field aligned approximately along the [010] direction of the epitaxial film. For coverages < 4 ML, no indication of ferromagnetism is observed. This suggests that the island growth of the interfacial region is

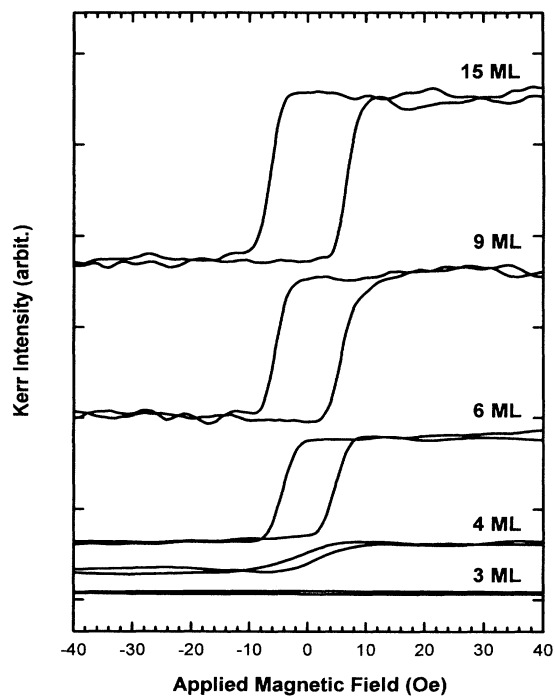


FIG. 3. SMOKE hysteresis loops for 3, 4, 6, 9, and 15 ML Fe overlayers. The applied magnetic field was aligned along the (010) direction.

sufficient to prevent the development of ferromagnetism. At an Fe coverage of 4 ML a clear ferromagnetic hysteresis loop can be observed. As the Fe coverage is increased further, the hysteresis loop continues to increase in strength. The coercivity of the Fe overlayer was found to vary with the thickness of the Fe film. The values were found to be approximately 3 Oe for 4–6 ML and 7 Oe for 8–20 ML Fe overlayers, respectively. These values are lower than the 25 Oe observed for 100–150 Å Fe overlayers grown on clean GaAs(100) [24]. This discrepancy is likely due to either the greater film thickness in Ref. [24] (reducing the quality of the overlayer) or to the effects of As interdiffusion in the Fe overlayer.

In summary, we have demonstrated a new procedure for the growth of Fe on S-passivated GaAs(100) which inhibits the diffusion of As through the overlayer by the S acting as a surfactant. An epitaxial bcc Fe(100) film can be grown, its surface being stabilized by the continual segregation of about 70% of the S in a $c(2 \times 2)$ ordered overlayer. The samples exhibit ferromagnetic behavior for Fe coverages >4 ML. The low coercive force of these films is evidence for high quality epitaxy.

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- [1] *Ultrathin Magnetic Structures*, edited by B. Heinrich and J. A. C. Bland (Springer-Verlag, Berlin, 1993), Volumes I and II.
 [2] J. R. Waldrop and R. W. Grant, *Appl. Phys. Lett.* **34**, 630 (1979).
 [3] S. A. Chambers, F. Xu, H. W. Chen, I. M. Vitomirov, S. B.

- Anderson, and J. H. Weaver, *Phys. Rev. B* **34**, 6605 (1986).
 [4] J. J. Krebs, B. T. Jonker, and G. A. Prinz, *J. Appl. Phys.* **61**, 2596 (1987).
 [5] R. F. C. Farrow, S. S. P. Parkin, and V. S. Speriosu, *J. Appl. Phys.* **64**, 5315 (1988).
 [6] E. Yablonovitch, C. J. Sandroff, R. Bhat, and T. Gmitter, *Appl. Phys. Lett.* **51**, 439 (1987).
 [7] M. S. Carpenter, M. R. Melloch, B. A. Cowans, Z. Dardas, and W. N. Delgass, *J. Vac. Sci. Technol. B* **7**, 845 (1989).
 [8] S. Gwo, K. J. Chao, A. R. Smith, C. K. Shih, K. Sadra, and B. G. Streetman, *J. Vac. Sci. Technol. B* **11**, 1509 (1993).
 [9] S. Matsumoto, S. Yamaga, and A. Yoshikawa, *Appl. Surf. Sci.* **60–61**, 274 (1992).
 [10] T. Scimeca, Y. Muramatsu, M. Oshima, H. Oigawa, Y. Nannichi, and T. Ohno, *J. Appl. Phys.* **71**, 4405 (1992).
 [11] H. Oigawa, J. F. Fan, Y. Nannichi, and M. Kawabe, *Jpn. J. Appl. Phys.* **29**, L544 (1990).
 [12] Z. H. Lu, M. J. Graham, X. H. Feng, and B. X. Yang, *Appl. Phys. Lett.* **62**, 2932 (1993).
 [13] H. Xia, W. N. Lennard, G. R. Massoumi, J. J. J. van Eck, L. J. Huang, W. M. Lau, and D. Landheer (to be published).
 [14] M. Tanimoto, H. Yokoyama, M. Shinohara, and N. Inoue, *Jpn. J. Appl. Phys.* **33**, L279 (1994).
 [15] T. Ohno and K. Shiraishi, *Phys. Rev. B* **42**, 11 194 (1990).
 [16] T. Ohno, *Surf. Sci.* **255**, 229 (1991).
 [17] T. Scimeca, Y. Muramatsu, M. Oshima, H. Oigawa, and Y. Nannichi, *Phys. Rev. B* **44**, 12 927 (1991).
 [18] G. W. Anderson, M. C. Hanf, and P. R. Norton (to be published).
 [19] K. O. Legg, F. Jona, D. W. Jepsen, and P. M. Marcus, *Surf. Sci.* **66**, 25 (1977).
 [20] K. O. Legg, F. Jona, D. W. Jepsen, and P. M. Marcus, *J. Phys. C* **10**, 937 (1977).
 [21] W. F. Egelhoff, Jr., *Solid State Mater. Sci.* **16**, 213 (1990).
 [22] S. Tanuma, C. J. Powell, and D. R. Penn, *Surf. Inter. Anal.* **11**, 577 (1988).
 [23] S. D. Bader, *J. Magn. Magn. Mater.* **100**, 440 (1991).
 [24] J. M. Florczak and E. D. Dahlberg, *Phys. Rev. B* **44**, 9338 (1991).