Evolution of the surface morphology of Fe grown on GaAs (100), (311)A, and (331)A substrates by molecular beam epitaxy

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We study the growth of Fe by molecular beam epitaxy on GaAs (100), (311)A, and (331)A substrates in dependence on the termination (reconstruction) of the GaAs surface and Fe growth temperature. Crystal quality and surface morphology of the 20- and 160-nm-thick Fe layers are characterized by double-crystal x-ray diffraction and atomic force microscopy. On GaAs (100) substrates we obtain very smooth Fe layers for As-rich surface reconstructions at a growth temperature of 50 °C. Less As-rich surface reconstructions produce macroscopic defects whose density increases on more Ga-rich surface reconstructions. On GaAs (311)A and (331)A substrates smooth layers with good crystal quality are obtained at 0 °C. The high density of macroscopic defects in these Fe layers is again eliminated on As-saturated surfaces. The evolution of the Fe surface morphology on the micron-length scale and the successful elimination of macroscopic defects on As-saturated GaAs substrates is highly relevant for application of these layers, in particular, their integration with the unique lateral semiconductor nanostructures formed on high-index GaAs (311)A and (331)A substrates. © 2001 American Institute of Physics.

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I. INTRODUCTION

Fe-on-GaAs is one of the most widely investigated epitaxial ferromagnetic metal/semiconductor hybrid system. This is not only because GaAs provides an ideal substrate for the epitaxial growth of Fe and related metallic multilayers with respect to crystal structure and lattice mismatch, but also due to the increasing interest in integrating magnetic thin films with semiconductor heterostructures for the realization of spin-sensitive devices.² Fe, with its bulk lattice constant of approximately half that of zinc-blende GaAs (\(2a_{\text{Fe}}/a_{\text{GaAs}} = 1.012\)) grows epitaxially in the stable (bcc) phase on GaAs (100) and (110) substrates by molecular beam epitaxy (MBE).³ The magnetic properties, e.g., the evolution of ferromagnetic order and the relationship between uniaxial and cubic magnetic anisotropy, as a function of layer thickness as well as the magnetic reversal processes have been investigated in detail.⁴⁻⁶ They have been related to interfacial compound formation and the atomic-scale nucleation processes imaged by electron diffraction and scanning tunneling microscopy (STM) for various Ga- and As-terminated GaAs (100) substrate surface reconstructions.⁷⁻⁹

On the other hand, very little information is available on the micron-scale surface morphology of Fe layers grown on GaAs substrates and the appearance of macroscopic defects which is highly relevant in view of lithographic patterning and local magnetic properties for device fabrication and reliability.

In the present article we study the growth of Fe by MBE on GaAs (100), (311)A, and (331)A substrates to obtain macroscopically smooth layers. The crystal quality and surface morphology of the 20- and 160-nm-thick Fe layers are characterized by double-crystal x-ray diffraction (XRD) and atomic force microscopy (AFM) in dependence on the growth temperature and termination (reconstruction) of the GaAs surface. As-terminated (2 × 4) reconstructed GaAs (100) surfaces, commonly established during MBE of GaAs, produce smooth Fe layers when grown at 50 °C. The Fe layers, however, exhibit a large density of macroscopic defects. The density of these macroscopic defects strongly increases for growth on the Ga-terminated (4 × 6) reconstructed GaAs (100) surface. Complete elimination of the macroscopic defects is achieved through growth on more As-rich (2 × 1) and (4 × 4) reconstructed GaAs (100). On the Ga-terminated GaAs (311)A and (331)A substrate smooth layers are obtained at 0 °C. The density of macroscopic defects, however, is even larger compared to that on Ga-terminated GaAs (100) substrates. Saturation with As effectively eliminates the defects also on these high-index substrates.

II. SAMPLE PREPARATION

The Fe layers were grown in an As-free metal MBE chamber connected to the preparation chamber of a three chamber III–V semiconductor MBE machine. Growth of the 70-nm-thick GaAs buffer layers was performed on semi-insulating GaAs (100), GaAs (311)A, and GaAs (331)A substrates at optimized growth conditions (growth temperature between 580 and 620 °C, growth rate 0.2 μm/h, and group V–III ratio of about 5) before establishing the different Ga- and As-terminated surface reconstructions monitored by reflection high-energy electron diffraction (RHEED): On GaAs (100) the As-rich (2 × 1) surface reconstruction is obtained when cooling the substrate down to 400 °C under As flux.
while the (4×4) surface reconstruction is established when, after stopping the As flux at 500 °C, several Ga monolayers are deposited. The As-terminated (2×4) surface reconstruction is obtained when the As flux is reduced by a factor of 20 at the growth temperature and several Ga monolayers are deposited before cooling. The Ga-rich (4×6) surface reconstruction is stabilized when the As flux is stopped after growth and several Ga monolayers are deposited before cooling. The As-saturated GaAs (311)A and (331)A surfaces are prepared in an analogous manner to the stabilization of the (2×1) GaAs (100) surface reconstruction with, however, no distinct modifications of the surface structure observable by RHEED. After transfer into the metal MBE chamber the actual reconstructions of the GaAs surfaces were again confirmed by RHEED before the Fe growth was started. The growth rate of Fe was 0.2 nm/min, and the growth temperature was between 0 and 175 °C with the background pressure in the MBE chamber of 1×10⁻¹⁰ Torr. The structural and morphological properties of the Fe layers with thicknesses of 20 and 160 nm were characterized in situ by RHEED and directly after removing the samples from the MBE growth chamber by AFM in air. For high-resolution x-ray studies (theta–two-theta scans) the Fe layers were capped with a 8-nm-thick protective Ag layer grown at 0 °C.

## III. EXPERIMENTAL RESULTS

### A. Fe on GaAs (100)

Figure 1(a) shows the AFM image of a 20-nm-thick Fe layer grown on a (2×4) reconstructed GaAs (100) substrate at 175 °C. The surface morphology exhibits rather smooth rectangular-shaped islands with micron side lengths. Deep holes are between these islands resulting in an overall rough surface morphology. This is evident also from the broad RHEED streaks during Fe growth confirming the absence of long range order of the surface morphology as reported previously. A number of macroscopic defects, several microns apart with micron diameters is visible in large-scale images. Table I summarizes the structural data of the Fe (100) layers discussed here. Very smooth Fe layers are obtained when grown at 50 °C [see the AFM image in Fig. 1(b) of the 20-nm-thick Fe layer]. The surface roughness amounts to several monolayers over micron distances. This indicates quasi two-dimensional Fe growth by nucleation and coalescence of a dense, homogeneous distribution of nanometer-scale islands due to the reduced Fe migration length. A larger growth rate is found to support this behavior at higher growth temperature. Narrow RHEED streaks during Fe growth, similar to those from GaAs agree with the existence of long range order and high structural perfection of the growing surface. The number of macroscopic defects, one seen in the AFM scan field in Fig. 1(b) as bright (high) area, however, is not reduced.

When the first 10 monolayers of Fe are grown at 50 °C and the substrate temperature is then raised to 100 or 175 °C for growth of the 20-nm-thick layer [Fig. 1(c)], the surface roughens after increase of the substrate temperature and develops a high density of small holes. Macroscopic defects are present with similar densities as in Fe layers grown at 50 °C. The formation of rectangular micron-sized islands obtained for entire growth at 175 °C, however, is not observed due to the quasi two-dimensional growth at 50 °C of the first 10 Fe monolayers.

Further lowering the substrate temperature to 0 °C results in very rough Fe layers, most probably due to a too small surface migration length [Fig. 1(d)], with a similar density of macroscopic defects. Hence, the optimum growth temperature for obtaining smooth Fe layers on GaAs (100)

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**Table I.** rms roughness determined from a 2×2 μm² AFM scan field and density of macroscopic defects of the 20-nm-thick Fe layers on GaAs (100) substrates in dependence on the surface reconstruction and growth temperature. The defect density is determined from 100×100 μm² images by optical microscopy.

<table>
<thead>
<tr>
<th>GaAs (100) surface reconstruction</th>
<th>Fe growth temperature in °C</th>
<th>Fe layer rms roughness in nm</th>
<th>Defect density in 10 000 μm⁻²</th>
</tr>
</thead>
<tbody>
<tr>
<td>(2×4)</td>
<td>Fig. 1(a)</td>
<td>175</td>
<td>10</td>
</tr>
<tr>
<td>(2×4)</td>
<td>Fig. 1(b)</td>
<td>50</td>
<td>15</td>
</tr>
<tr>
<td>(2×4)</td>
<td>Fig. 1(c)</td>
<td>50/175</td>
<td>20</td>
</tr>
<tr>
<td>(2×4)</td>
<td>Fig. 1(d)</td>
<td>0</td>
<td>20</td>
</tr>
<tr>
<td>(4×6)</td>
<td>Fig. 3(a)</td>
<td>50</td>
<td>300</td>
</tr>
<tr>
<td>(2×1)</td>
<td>Fig. 3(b)</td>
<td>50</td>
<td>0</td>
</tr>
</tbody>
</table>

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substrates is around 50 °C. This growth temperature is also beneficial for suppression of the formation of interface compounds between Fe and GaAs which have reduced magnetic moment and/or absence of ferromagnetic order.11

Figure 2 shows the XRD spectra of 20- and 160-nm-thick Fe layers grown at (a) and (c) 50 °C and (b) and (d) 175 °C in the range between the symmetric GaAs (400) and (200) reflections. In addition to the Fe (200) reflection, in (b) and (d) weak reflections from the antiferromagnetic Fe3As phase and the Fe2Ga3−xAsx compound with reduced magnetic moment12 formed at the Fe–GaAs interface are clearly seen, which are not detectable in (a) and (c). The width of the Fe (200) reflection is comparable for all layers revealing similar crystal perfection. It is interesting to note the slightly different degree of relaxation of the Fe layers depending on the presence of the (Fe, Ga)As interface layer and/or growth temperature and the Fe layer thickness with the lattice mismatch of 1.2% between bulk GaAs and Fe relaxed to about 0.8% in the 160-nm-thick Fe layers.

To avoid the formation of (Fe, Ga)As interface compounds which deteriorate the ferromagnetic order at the Fe–GaAs interface we prepared the Ga-terminated (4×6) reconstructed GaAs (100) surface prior to Fe growth. Growth of Fe on this surface reconstruction has been reported to fully recover the ferromagnetic order at the Fe–GaAs interface.13 However, the density of macroscopic defects increases by about one order of magnitude on the Ga-terminated surface as shown in the AFM image of Fig. 3(a) of the 20-nm-thick Fe layer grown at 50 °C. Entire elimination of the defects is achieved on the (2×1) and (4×4) reconstructed As-rich GaAs (100) surfaces with the very smooth Fe surface morphology for growth at 50 °C [see Fig. 3(b), AFM image of 20 nm Fe on (2×1) GaAs (100)]. The surface morphology for Fe on (4×4) GaAs (100) is indistinguishable. Irrespective of the density of macroscopic defects the XRD spectra equal that of Fig. 2(a).

FIG. 2. Double-crystal XRD spectra of (a) and (b) 20- and (c) and (d) 160-nm-thick Fe layers grown at (a) and (c) 50 °C and (b) and (d) 175 °C on GaAs (100) substrates with As-terminated (2×4) surface reconstruction recorded in the range between the symmetric GaAs (400) and (200) reflections. The diffraction peaks due to the Fe layer and the interface compounds in (b) and (d) are indicated. The Ag peak is from the cap layer.

FIG. 3. AFM images of 20-nm-thick Fe layers grown at 50 °C on GaAs (100) substrates (a) with Ga-terminated (4×6) and (b) with As-rich (2×1) surface reconstructions. Table I summarizes the structural data of the Fe layers.
IV. DISCUSSION

The surface morphology of GaAs \(\{100\}, \{311\}A, \) and \(\{331\}A\) has been studied extensively by AFM and other methods in our laboratory for various growth conditions in particular those producing macroscopic defects in the Fe layers proving the absence of macroscopic defects or any correlated features already on the starting GaAs surfaces.\(^14\)–\(^16\) Moreover, the surprising relationship of the appearance and density of macroscopic defects with the termination of the GaAs surface clearly excludes any specific Fe growth conditions as their origin, but instead, independent of the GaAs surface orientation and preparation, a common Fe–GaAs interface formation mechanism during the initial growth stage: Recent studies of Fe grown on GaAs \(\{100\}\) substrates with different As-terminated surface reconstructions have pointed towards the formation of a common interface structure based on Fe bonded to a full plane of As. The Fe layers showed the same magnetic properties though the atomic scale nucleation processes imaged by STM were quite different.\(^8\) Moreover, x-ray photoelectron spectroscopy revealed outdiffusion of Ga and As into the Fe layer, with the respective amount making available the first full As plane. Hence, we assume, the As-saturated GaAs surface is preferred for growth of Fe on GaAs minimizing Ga outdiffusion, i.e., Fe–Ga exchange reactions at the interface which might enhance the probability to generate macroscopic defects. The density of macroscopic defects is thus directly connected with the surface termination and, thus, increases on less As-rich surface reconstructions. This agrees with the reported three-dimensional clustering of Fe on GaAs \(\{110\}\)\(^17\) compared to the layer-by-layer growth on GaAs \(\{100\}\).\(^10\) On GaAs \(\{110\}\) surfaces, Ga and As atoms are located in the same plane, not allowing the formation of a planar Fe–As interface. The growth on As-saturated GaAs substrates has, however, to be compromised with the reported suppression of nonferromagnetic interface layers for growth on Ga terminated \((4\times6)\) reconstructed GaAs \(\{100\}\) substrates discussed before.\(^13\) Fortunately, the lower optimum Fe growth temperature on the GaAs \(\{311\}A\) and \(\{331\}A\) substrates favors...

![Figure 4](image1.png)

**FIG. 4.** AFM images of 20-nm-thick Fe layers grown at 50 °C on (a) GaAs \(\{311\}A\) and (b) GaAs \(\{331\}A\) substrates, at 0 °C on (c) GaAs \(\{311\}A\) and (d) \(\{331\}A\) substrates, and at 0 °C on As saturated (e) GaAs \(\{311\}A\) and (f) GaAs \(\{331\}A\) substrates. Table II summarizes the structural data of the Fe layers.

**TABLE II.** rms roughness determined from a 2×2 \(\mu\)m\(^2\) AFM scan field and density of macroscopic defects of the 20-nm-thick Fe layers on GaAs \(\{311\}A\) and \(\{331\}A\) substrates in dependence on the surface termination and growth temperature. The defect density is determined from 100×100 \(\mu\)m\(^2\) images by optical microscopy.

<table>
<thead>
<tr>
<th>GaAs substrate orientation</th>
<th>({311}A)</th>
<th>({331}A)</th>
<th>({311}A)</th>
<th>({331}A)</th>
<th>({311}A; As)</th>
<th>({331}A; As)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe growth temperature in °C</td>
<td>50</td>
<td>50</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Fe layer rms roughness in nm</td>
<td>2.3</td>
<td>4.6</td>
<td>0.78</td>
<td>1.16</td>
<td>0.46</td>
<td>0.45</td>
</tr>
<tr>
<td>Defect density in 10 000 (\mu)m(^2)</td>
<td>630</td>
<td>1100</td>
<td>560</td>
<td>320</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

**FIG. 5.** Double-crystal XRD spectra of 160-nm-thick Fe layers grown at 0 °C on (a) GaAs \(\{311\}A\) and (b) GaAs \(\{331\}A\) substrates recorded in the vicinity of the asymmetric GaAs \(\{422\}\) reflection.
the suppression of these nonferromagnetic interface layers towards the direct integration of thin epitaxial Fe layers with the unique lateral semiconductor nanostructures formed on these high-index GaAs substrates.\textsuperscript{15,16}

V. CONCLUSION

We have investigated the MBE growth of Fe on GaAs (100), (311)A, and (331)A substrates in dependence on the growth temperature and termination of the GaAs surface. Very smooth Fe layers, free of macroscopic defects, are obtained on GaAs (100) substrates with As-rich surface reconstruction at 50 °C growth temperature. Less As-rich surface reconstructions produce macroscopic defects whose density increases with more Ga-rich surface reconstructions. At higher (175 °C) and lower (0 °C) growth temperatures the Fe layer roughens and consists of micron-sized islands for growth at 175 °C. A similar island morphology is formed on GaAs (311)A and (331)A substrates at 50 °C while smooth Fe layers with good crystal quality are obtained at 0 °C. The large number of macroscopic defects in these Fe layers is again eliminated with As-saturated surfaces. The successful growth of very smooth, epitaxial Fe layers, free of macroscopic defects, is of high relevance for applications and, in particular their integration with the unique lateral semiconductor nanostructures formed on high-index GaAs (311)A and (331)A substrates.

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