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Scaling behaviour and evolution of ferromagnetism in epitaxial Fe/GaAs(100) and Fe/InAs(100)

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Abstract

The scaling behaviour of the thickness evolution of ferromagnetism in single crystal Fe films on GaAs(100)-4 × 2 and InAs(100)-4 × 2, grown by molecular beam epitaxy, have been studied. In both systems there is a superparamagnetic-ferromagnetic transition whose onset occurs around 4.5 monolayers (ML) Fe. The susceptibility around the transition follows a scaling behaviour; $\chi \sim (1 - d/d_c)^{-\gamma}$ with $\gamma = 2.13 \pm 0.30$ for Fe/GaAs and $\gamma = 2.21 \pm 0.25$ for Fe/InAs, suggesting the 2D percolation of clusters. © 1999 Elsevier Science S.A. All rights reserved.

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1. Introduction

Of all the Fe/III–V semiconductor systems, Fe/GaAs(100) has received the most attention [1–6]. Fe/InAs(100) offers further opportunities for potential magneto-electronic applications in controlling electrical as well as magnetic properties. In contrast to GaAs, *I*–*V* measurements in the temperature range 2.5–304 K show that Fe forms a low resistance ohmic contact on InAs [7]. InAs also has a higher low-field mobility than both GaAs and InP, which makes it an excellent candidate for high speed field effect transistors [8,9]. We have found that despite the large lattice mismatch (–5.4%, compared with +1.4% for Fe/GaAs), Fe grows epitaxially on InAs(100) at both 170°C and at room temperature, as evidenced by LEED. It is also noteworthy that the sense of mismatch is different – Fe being expanded in-plane on InAs(100) which may help explain magnetic differences in the two systems. The magnetic hysteresis loops measured using in situ MOKE showed that thick Fe films display a cubic anisotropy with easy axis along $\langle 100 \rangle$, the easy axis of bcc Fe. However, in the ultrathin regime the magnetic properties of these two systems differ significantly. For example, the uniaxial anisotropy of Fe/InAs(100) is far weaker than in Fe/GaAs(100) and the uniaxial anisotropy easy axes are along different directions. In Fe/InAs(100) the uniaxial

easy axis is along [011], as opposed to $[0\bar{1}1]$ in Fe/GaAs(100) – further details are described elsewhere [10].

The results of equivalent studies on Co/Cu(100), a system known to exhibit layer-by-layer growth, above 2 ML, showed a paramagnetic-ferromagnetic transition which followed a 2D percolation [11]. A 2D percolation has also been observed for a system known to follow a 3D growth mode. Hope et al. have found a paramagnetic-ferromagnetic transition in Co/Cu(110) [12]. It is interesting to study the scaling behaviour in Fe/GaAs(100) and Fe/InAs(100) which also grow in 3D islands, but which are believed to display a superparamagnetic-ferromagnetic transition [13].

2. Experiments

This study was carried out in a ‘multiple-technique’ molecular beam epitaxy (MBE) system, which is described elsewhere [5,7,13] and includes in situ magneto-optical Kerr effect (MOKE) [14] and low energy electron diffraction (LEED). The Fe films were grown at a rate of approximately one monolayer (ML) per minute using an e-beam evaporator. The evaporator consists of an Fe rod, which is surrounded by a watercooled shield. The rod is heated directly by a filament and also by electron bombardment – rod held at around 700 V. The evaporation rate was measured by a quartz crystal oscillator and was calibrated using Fe/Ag(100) RHEED oscillations. The pressure during growth was around $7\text{--}8 \times 10^{-10}$ mbar. The MOKE experiments yield magnetisation–field (*M*–*H*) curves and give values which are proportional to the saturation and remanent

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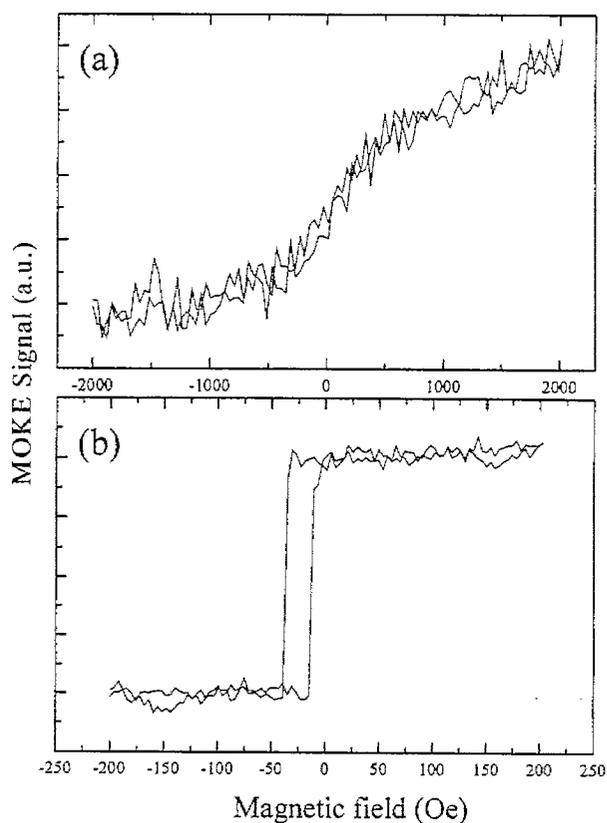


Fig. 1. In situ M - H (MOKE) signals for Fe/GaAs(100)- 4×2 with the magnetic field applied along $[0\bar{1}1]$. (a) Superparamagnetic signal from 3.0 ML Fe (single measurement). (b) Ferromagnetic hysteresis loop from 4.5 ML Fe (ten averages).

magnetisation. To enable the accurate measurement of MOKE intensity and coercive field with thickness, neither the sample nor the optical set-up was adjusted during the MOKE measurements. In these MOKE experiments the field was applied along the direction of the uniaxial anisotropy easy axis. Thus the field was applied along $[0\bar{1}1]$ for Fe/GaAs, whilst along $[011]$ for Fe/InAs.

The GaAs(100) substrates used in this study were commercial wafers upon which a buffer layer ($\sim 0.5 \mu\text{m}$) of homoepitaxial GaAs was grown to provide the smoothest possible surface and then As capped prior to removal from the semiconductor growth chamber. After transfer to the magnetic growth chamber the As cap layer was then desorbed by annealing. The As capping layer began to desorb at around 340°C and the substrate was further annealed to 550°C for 1 h to obtain a clean and ordered surface. All Fe films were grown at room temperature rather than higher temperature ($\sim 175^\circ\text{C}$) to reduce the intermixing of Fe with As at the interface. The InAs(100) substrates used were commercial wafers. Before loading into the UHV system, the substrates were cleaned using a combination of oxygen plasma etching and wet etching ($\text{HCl}/\text{H}_2\text{O} = 1:4$). After the substrate was annealed at 510°C for 30 min, clear LEED patterns were observed – this indicates that the cleaning procedure produces a well-

ordered surface. The fourth-order spots along the $[011]$ direction are very sharp and there is some streaking of the twofold spots in this direction, indicating an In-terminated 4×2 reconstruction of the surface [7,10].

3. Results and discussion

Fig. 1 shows MOKE data for Fe/InAs(100) before and after the transition. Fig. 1a shows a superparamagnetic M - H curve for 3.0 ML Fe – note that the curve is just becoming S-shaped and shows no hysteresis. The lack of hysteresis is consistent with either superparamagnetism [15], or 2D paramagnetism [16]. However, the M - H curves which were observed in this region are generally consistent with the Langevin function used to describe the magnetization of superparamagnetic clusters [15]. Fitting the M - H curves with a Langevin function, in the range of ± 1 kOe, the average value of the effective magnetic moment per cluster on Fe/GaAs(100)- 4×2 is estimated to be $(1.75 \pm 0.15) \times 10^4 \mu_B$, after 3 ML deposition. Assuming a bulk moment for the Fe and an island height of 5.0 ML after 3.0 ML deposition, the size of clusters maybe estimated. For this case the average island area is estimated to be 3400 \AA^2 ($58 \times 58 \text{ \AA}$). Using STM, Thibada et al. [17] found island sizes of $35 \times 90 \text{ \AA}$ (3150 \AA^2) for 1.0 ML of Fe on GaAs(001)- 2×4 . STM measurements on Fe/InAs(100) [7] have allowed us to measure the feature size directly and these are comparable with those deduced by fitting the M - H curves using the Langevin function. Further evidence supporting the superparamagnetic transition is that a bcc Fe LEED pattern was never observed at an Fe thickness less than that at which the first hysteretic M - H loops appear. This indicates that, in the region before the onset of ferromagnetism, the surface is not fully covered with Fe.

With further coverage, the islands coalesce and long range ferromagnetic ordering develops. The hysteresis loops after the onset of the ferromagnetic phase show that the films have a well defined magnetic coercivity and remanance ratio, indicating the behaviour of a continuous film. Fig. 1b shows such a ferromagnetic loop for 4.5 ML Fe. The M - H curves measured along the easy axes are initially linearly proportional to applied field. With increasing coverage they become S-shaped (with no hysteresis), becoming gradually S-shaped (with increasing hysteresis) and then becoming square with distinct hysteresis. This progression is indicative of superparamagnetic-ferromagnetic behaviour. The results of equivalent studies on Co/Cu(100) [11] showed a paramagnetic-ferromagnetic transition which differed to the present case in that there was a far more sudden change from S-shaped curves (with no hysteresis), to square hysteresis loops as the thickness was increased (i.e. no visible region of hysteretic S-shaped curves).

The susceptibility (χ) is defined as the derivative of the magnetisation with respect to the applied field, evaluated at

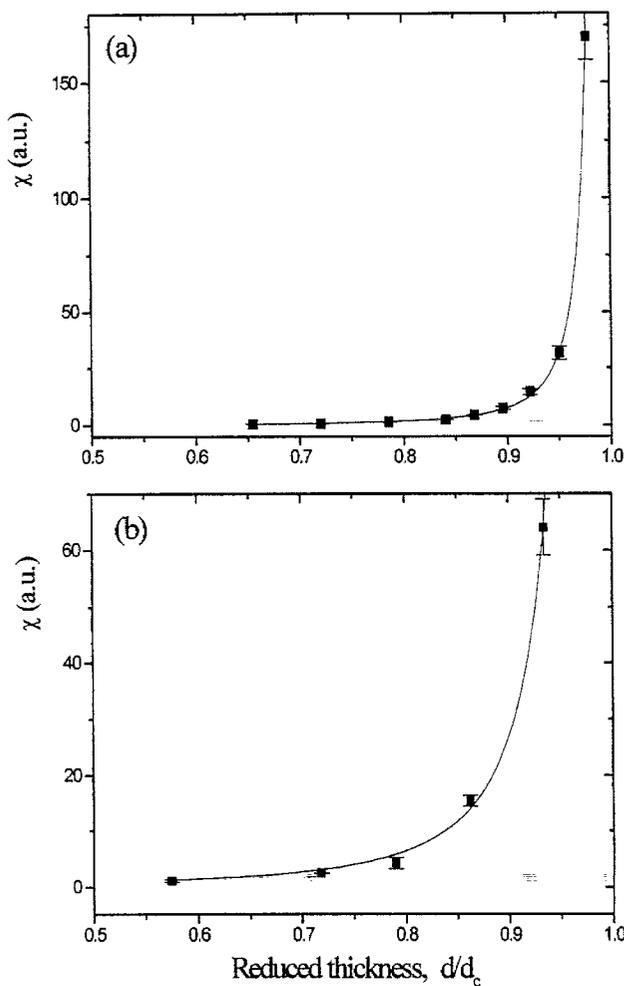


Fig. 2. Evolution of χ as a function of reduced thickness (d/d_c). The lines shown are guides to the eye using critical exponent values determined from Fig. 3. (a) Fe/GaAs(100) and (b) Fe/InAs(100).

zero field (Eq. (1)). A value which is directly proportional to the susceptibility has been determined by fitting the gradient of the superparamagnetic $M-H$ curves in the linear region (see Fig. 1a). By assuming that the percentage of Fe coverage on the substrate is directly proportional to the thickness of deposited Fe, we can apply a power law model (see Eq. (2)) in analysing the thickness induced transition in the susceptibility data. The divergence of χ occurs in a very narrow thickness range – at the critical thickness (d_c) it becomes infinite.

$$\chi = \left(\frac{\partial M}{\partial H} \right)_{H=0} \quad (1)$$

$$\chi \propto \left(1 - \frac{d}{d_c} \right)^{-\gamma} \quad (2)$$

Fig. 2 displays the susceptibility variation of the two systems in the region before the onset of ferromagnetism. Fig. 3 displays the susceptibility results of these data sets as $\log(\chi)$ versus $\log(1 - d/d_c)$ from which the critical expo-

nent, γ , has been determined by least squares fitting. The value of γ was determined from the slope of the log–log plot, by allowing d_c to vary until the fit parameters were optimised. The values obtained were $d_c = 3.82 \pm 0.05$ ML, $\gamma = 2.13 \pm 0.30$ for Fe/GaAs and $d_c = 3.48 \pm 0.05$ ML, $\gamma = 2.21 \pm 0.25$ for Fe/InAs.

Percolation theory regards the connectivity of randomly occupied lattice sites. As the number of occupied sites increases, clusters are formed and at a critical concentration of 0.59 [18] there is a finite probability of a cluster extending across the entire area of the lattice. The percolation model predicts γ values of 43/18 (2.389) and 1.66, for two dimensions (2D) and 3D, respectively. Clearly our results, for both Fe/GaAs and Fe/InAs are consistent with this 2D model.

Though the semiconductor surface is not isotropic due to the reconstruction and presence of dimer ridges [19,20], the lattice sites able to accommodate Fe atoms are regularly spaced and in principle, may be randomly occupied. Due to the relative sizes of the surface energies it is energetically favourable for the Fe atoms to aggregate together until the substrate is covered (Volmer–Weber mode). Hence, at first

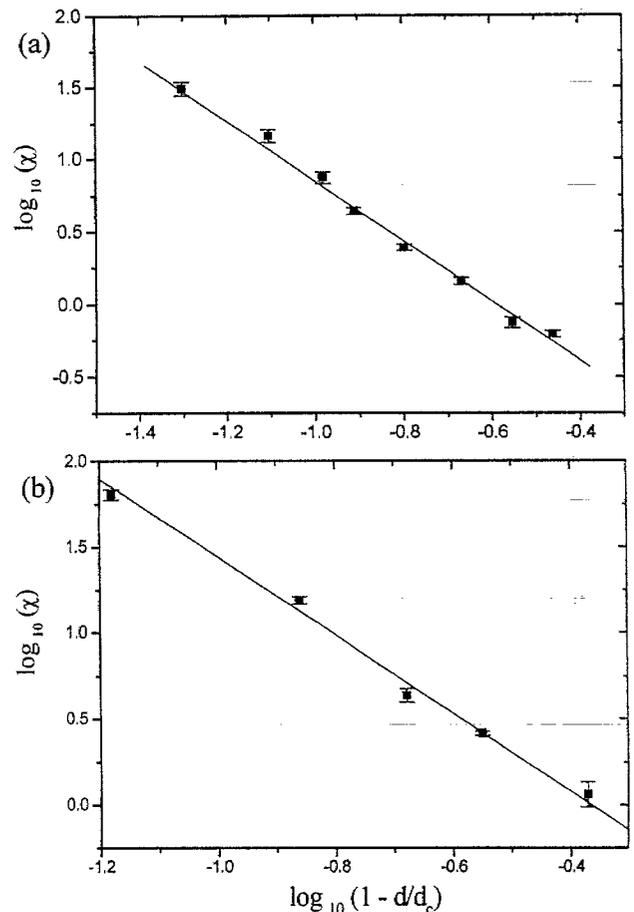


Fig. 3. Log–log plots of the data shown in Fig. 2. In each case the slope of the linear fit gives the critical exponent, γ . (a) Fe/GaAs(100) and (b) Fe/InAs(100).

sight it is surprising that the scaling behaviour seems to be that for a 2D percolation transition. In our experiment we have little control on exactly where the deposited atoms both land and actually lodge on the surface. The landing sites of the Fe atoms are believed to be random. However, due to the relative sizes of the surface energies, the Fe growth mode is island-like (3D) on both GaAs and InAs – thus the final resting sites of individual atoms are far from random. However, the transition observed does not concern individual atoms, but is believed to involve clusters of Fe atoms and it is these clusters which are randomly located on the surface. Hence, the superparamagnetic–ferromagnetic transition is consistent with a 2D percolation of Fe clusters on the semiconductor surface.

4. Conclusions

We have successfully grown epitaxial Fe on both GaAs(100) and InAs(100). We have found that the susceptibility follows a scaling behaviour with critical exponent $\gamma = 2.13 \pm 0.30$ for Fe/GaAs(100) and $\gamma = 2.21 \pm 0.25$ for Fe/InAs(100). These values are consistent with a 2D percolation transition. Since the growth of Fe is three-dimensional in nature and atoms aggregate together, it is somewhat surprising that our results fit a 2D percolation model which relies on the statistical occupation of sites. This paradox may be explained by considering that the observed transition is probably a 2D percolation of clusters arranged randomly on the surface, rather than a 2D percolation of randomly arranged individual atoms.

Whilst there are differences in the magnetic properties of these two systems (namely the strength and easy direction of the uniaxial magnetic anisotropies), this work highlights certain similarities. Firstly, that Fe forms 3D islands on both substrates. Secondly, both Fe/GaAs(100)- 4×2 and Fe/InAs(100)- 4×2 undergo a distinct transition in the

susceptibility behaviour at almost the same Fe thickness, which in each case is explained by a 2D percolation of clusters; although both systems follow a 3D-island growth in the initial stages.

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