

Interface atomic structure and magnetic anisotropy in ultrathin Fe films grown by thermal deposition and pulsed laser deposition on GaAs(001)

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Thermal deposition (TD) and pulsed laser deposition (PLD) techniques were used to prepare GaAs/Fe(001) crystalline ultrathin films. The magnetic environment of Fe at the Fe/GaAs(001) interface was investigated by means of conversion electron Mössbauer spectroscopy using an ⁵⁷Fe probe layer having the equivalent thickness of two atomic layers. The magnetic anisotropies were measured using ferromagnetic resonance. PLD samples were affected by an appreciably higher degree of interface alloying compared with the TD films. The interface in-plane fourfold (cubic) anisotropy is significantly increased and the in-plane interface uniaxial anisotropy is significantly decreased in the PLD samples compared to those prepared using TD. The effective perpendicular demagnetizing field in the PLD films is significantly decreased compared to the TD samples due to an unexpected increase in the interface uniaxial perpendicular anisotropy at the Fe/Au(001) interface. © 2007 American Institute of Physics. [DOI: [10.1063/1.2711071](https://doi.org/10.1063/1.2711071)]

Introduction. This paper has been motivated by the ongoing discussion of the structural and chemical properties of crystalline GaAs/Fe(001) interfaces.^{1,2} It is directed towards two goals: (a) Understanding the Fe chemical environment at the GaAs/Fe(001) interfaces which were prepared by the thermal deposition (TD) and pulse laser deposition (PLD) techniques and (b) understanding the role of TD and PLD on the interface magnetic anisotropies in GaAs/Fe(001) based structures.

Growth Studies. The Fe films were deposited on a commonly used 4 × 6-GaAs(001) reconstructed template. The 4 × 6 surface reconstruction was obtained by annealing the GaAs wafer at ≈600 °C following the hydrogen cleaning and Ar⁺ sputtering. Scanning tunneling microscopy (STM) images indicate that this reconstruction consists of two mixed phases:³ (a) a 2 × 6 reconstruction having As dimers and dangling bonds oriented along the [1̄10] crystallographic direction and (b) a 4 × 2 GaAs(001) reconstruction terminated by Ga with the Ga dimers oriented along the [110] crystallographic orientation. The 4 × 6 reconstruction is clearly visible in reflection high energy electron diffraction (RHEED) studies, but the STM images revealed that the 2 × 6 reconstruction occupies 95% of the surface. The Fe

growth was carried out at room temperature (RT). The Fe nucleates along the rows of As dimers.¹ After deposition of the equivalent three atomic layers, a continuous film is formed having atomic terraces approximately 3–4 nm wide on average. The equivalent of two atomic layers of ⁵⁷Fe was used as a probe layer for Mössbauer spectroscopy. They were deposited using TD and PLD at the GaAs(001) interface and inside the Fe film. The studies in this paper were carried out on the following structures: GaAs/2⁵⁷Fe_{PLD}/8Fe_{TD}/20Au(001), GaAs/2⁵⁷Fe_{PLD}/8Fe_{PLD}/20Au(001), GaAs/2⁵⁷Fe_{TD}/8Fe_{TD}/20Au(001), GaAs/5Fe_{TD}/2⁵⁷Fe_{TD}/3Fe_{TD}/20Au(001), and GaAs/7Fe_{TD}/2⁵⁷Fe_{TD}/7Fe_{TD}/20Au(001). The integers represent the number of atomic layers, and the subscripts describe the deposition technique.

Mössbauer Studies. The magnetic states of Fe atoms at the GaAs/Fe interface were identified using conversion electron Mössbauer spectroscopy (CEMS) at RT. The spectra are shown in Fig. 1. The relative contributions of the spectrum components and the corresponding hyperfine fields (H_f) are summarized in Fig. 2. The Mössbauer spectra for the TD and PLD films show a broad hyperfine field distribution for the ⁵⁷Fe probe layer grown directly on the GaAs(001) template. Eight to ten Mössbauer components were required to account for the measured spectra. The multicomponent fitting was constrained by the following procedure: (a) the highest mea-

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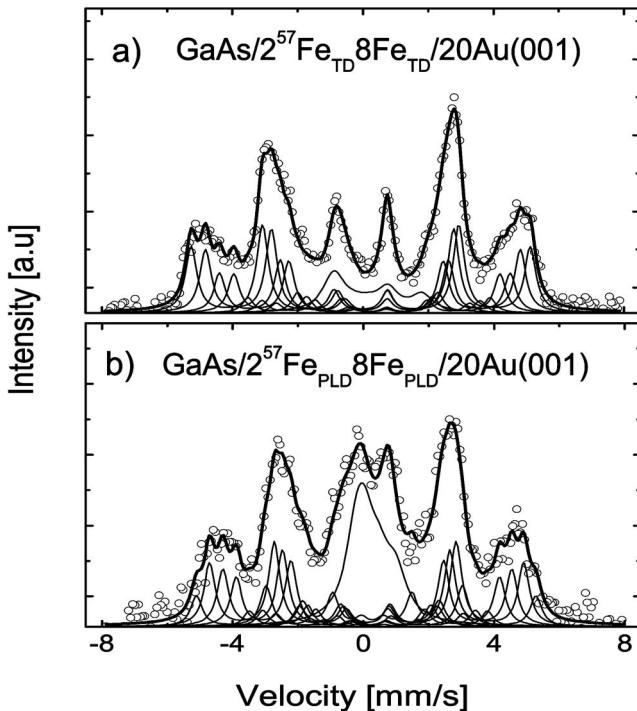


FIG. 1. Conversion electron Mössbauer spectra (CEMSs) for (a) TD and (b) PLD samples. The thin solid lines represent the individual components of Mössbauer spectra corresponding to the different chemical environments of the Fe. The thick solid line represents the overall fit to the data.

sured hyperfine field was fixed at 322 kOe obtained from the GaAs/5Fe_{TD}/2⁵⁷Fe_{TD}/3Fe_{TD}/20Au(001) sample at RT (see discussion of Fig. 3) and (b) hyperfine field H_f was expected to decrease by ~ 23 kOe on average for each replacement of the nearest neighbor (NN) Fe atom with either an As or a Ga atom. The main difference between the TD and PLD samples was observed at the lowest range and at the highest range of the H_f components, otherwise the components between these extremes were remarkably similar. For the TD film the lowest $H_f \approx 86$ kOe and represents only 11% in the relative intensity (RI), while for the PLD film $H_f \approx 38$ kOe and RI = 22%. Clearly, the high kinetic energy of the PLD Fe clusters leads to a strong incorporation of Fe into the subsurface of the GaAs lattice. The relative intensities for the components with the highest hyperfine fields (above 298 kOe) were found to be 47% and 27% for the TD and PLD films, respectively. Note that for the TD sample, 47% represents nearly a

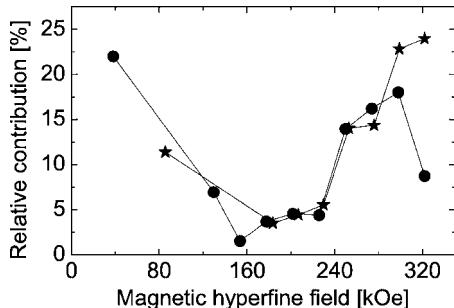


FIG. 2. Relative contribution of the spectrum components vs the corresponding magnetic hyperfine fields from the data in Fig. 1. (●) and (★) represent the PLD and TD samples, respectively.

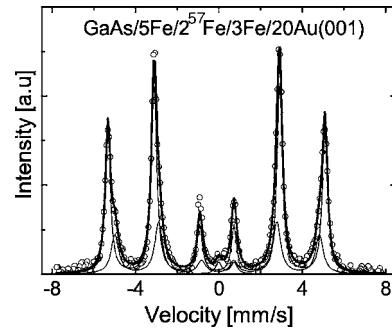


FIG. 3. Conversion electron Mössbauer spectra (CEMSs) for GaAs/5Fe/2⁵⁷Fe/3Fe/20Au(001) film.

whole atomic layer. Therefore the atoms in this layer are surrounded by seven to eight NN of Fe. This again supports the view that the TD samples have a better interface lattice structure than those deposited by means of PLD. The lowest hyperfine field components correspond to Fe states having mostly a nonmagnetic atomic environment. This is in agreement with the recent STM images¹ indicating that during deposition some of the Fe penetrates into the GaAs(001) substrate. These Fe atoms substitute for Ga atoms in the second layer, displacing them into an interstitial position. In addition, it is suggested that the Fe atoms inside the top As layer together with the top Fe layer form Fe₂As compound seed crystals for the bcc Fe growth. The observed multitude of hyperfine field components supports this view of a complex transition from a perfect GaAs substrate to an Fe film unaffected by interface mixing. Even the TD GaAs/5Fe/2⁵⁷Fe/3Fe/20Au(001) film where the probe layer is located five atomic layers away from the GaAs surface requires the two component fit, see Fig. 3: (a) $H_f = 322$ kOe and RI=72%; (b) $H_f = 304$ kOe and RI=28%. The $H_f = 304$ component represents a state with seven Fe atoms in the NN configuration. This state has not been observed in the growth of Fe on Ag(001) substrates that were cut from a single crystalline Ag boule.⁴ There is an essential difference between these two structures. During the growth of Fe on GaAs(001), the interface As acts partly as a surfactant and the equivalent of 0.6 monolayer (ML) of As floats on top of the Fe film. Ag in the Ag/Fe(001) structure also acts as a surfactant but it is known that it is definitely not incorporated into the Fe film. The component with $H_f = 304$ kOe in the GaAs/5Fe/2⁵⁷Fe/3Fe/20Au(001) sample indicates that some of the surfactant As is left inside the Fe film. This trend decreases with the film thickness. In fact in GaAs/7Fe/2⁵⁷Fe/7Fe/20Au(001) the two components have the following parameters: (a) $H_f = 326$ kOe and RI=82%; (b) $H_f = 310$ kOe and RI=18%. Obviously the second sample is much closer to the ideal bulk. Somewhat lower values of H_f at RT compared to the bulk H_f (331 kOe) are expected. Ultrathin films have a lower critical point than bulk Fe, and H_f strongly depends on the film thickness.

Ferromagnetic Resonance Studies. The results of ferromagnetic resonance (FMR) measurements have shown that differences in the Fe interfaces between the TD and PLD samples strongly affect the magnetic anisotropies, see Table I and Fig. 4. The magnetic free energy density for the in-plane

TABLE I. Samples and their measured magnetic properties.

Sample	$K_{1,\text{eff}}[10^5 \text{ erg/cm}^3]$	$K_{u,\parallel}[10^5 \text{ erg/cm}^3]$	$4\pi M_{\text{eff}}[\text{kG}]$
GaAs/2 ⁵⁷ Fe _{TD} /8Fe _{TD} /20Au(001)	2.02	-7.01	14.74
GaAs/2 ⁵⁷ Fe _{PLD} /8Fe _{PLD} /20Au(001)	-0.43	-2.25	9.60
GaAs/2Fe ⁵⁷ _{PLD} 8Fe _{TD} /20Au(001)	0.73	-1.72	13.92

oriented saturation magnetization is given by the following expression:

$$K_{1,\text{eff}} \frac{\sin^2(2\varphi)}{4} + K_{u,\parallel}(\mathbf{n} \cdot \mathbf{m})^2 + 2\pi M_{\text{eff}}^2 \cos^2(\vartheta), \quad (1)$$

where \mathbf{m} is a unit vector in the direction of the saturation magnetization, $K_{1,\text{eff}}$ is the in-plane fourfold (cubic) anisotropy, $K_{u,\parallel}$ is the in-plane uniaxial anisotropy, \mathbf{n} is the direction of the in-plane uniaxial axis, φ is the azimuthal angle of the magnetization vector with respect to the [100] axis, ϑ is the polar angle of the magnetization with respect to the [001] axes, and $4\pi M_{\text{eff}}$ is the effective demagnetizing field perpendicular to the surface. The subscript eff indicates that the magnetic anisotropies in ultrathin films are composed of both bulk and interface contributions.⁶ Fe films prepared by TD on GaAs(001) are known to be well described by the superposition of bulk and interface anisotropies.^{5,7} The in-plane uniaxial anisotropy $K_{u,\parallel}$ is caused predominantly by the GaAs/Fe interface. It is inversely proportional to the film thickness, with the hard magnetic axis oriented along the [1̄10] crystallographic direction.³ PLD affects all interface magnetic anisotropies. The in-plane uniaxial anisotropy is significantly decreased by a factor of 4, see table and Fig. 4,

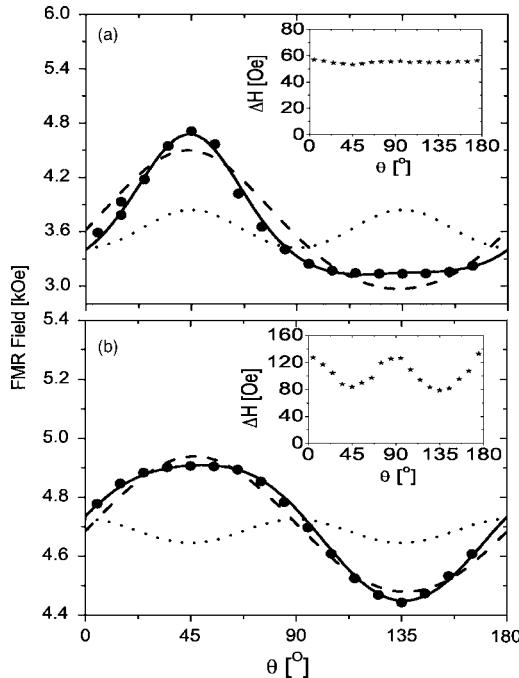


FIG. 4. FMR field at $f=24$ GHz as a function of the in-plane angle θ of dc field with respect to the [100] axis. GaAs(001)/10Fe/20Au(001) samples were prepared by (a) TD and (b) PLD. The dashed and dotted lines are the calculated resonance field contributions from $K_{u,\parallel}$ and $K_{1,\text{eff}}$, respectively. The solid lines represent an overall fit. The insets show the corresponding angular dependence of the FMR linewidth, ΔH .

and the interface fourfold anisotropy (negative with respect to the bulk K_1) is increased in magnitude and results in a negative $K_{1,\text{eff}}$ even for a 10 ML thick Fe film. $4\pi M_{\text{eff}}$ decreased by less than 1 kG in the GaAs/2Fe_{PLD}8Fe_{TD}/20Au(001) sample, whereas in the GaAs/10Fe_{PLD}/20Au(001) structure, $4\pi M_{\text{eff}}$ decreased by 5 kG. This means that the interface uniaxial perpendicular anisotropy is mostly increased at the Fe/Au(001) interface. The thickness dependence has shown that the total interface uniaxial anisotropy was increased from 0.9 erg/cm^2 for the TD films to 1.4 ergs/cm^2 for the PLD films. PLD also increases the magnetic damping. Magnetic damping in Fe films prepared by TD can be described by the Gilbert damping mechanism, see Ref. 8. The Gilbert damping is independent of the orientation of the magnetic moment. The magnetic damping in PLD samples is angular dependent, see insets in Fig. 4, with the maximum damping along the ⟨100⟩ crystallographic axes. The additional damping in PLD samples is caused by two magnon scattering.⁸ This implies that a network of lattice defects created by PLD is accompanied by a network of in-plane uniaxial anisotropies with the uniaxial anisotropy axes oriented along the ⟨100⟩ crystallographic axes.

Conclusions. CEMS has shown that the magnetic state of Fe at the GaAs(001) interface is very sensitive to the deposition technique. Fe films prepared by PLD exhibit a more intermixed interface than those prepared by thermal deposition. Consequently the Fe lattice in the PLD films can be expected to be clamped more strongly to the bcc GaAs(001) lattice than is the case for the TD films. The recent observation of interface shear² at the GaAs/Fe(001) interface suggests that the in-plane interface uniaxial anisotropy can be explained by a magnetoelastic term due to lattice shear.⁹ A significant decrease in the in-plane interface uniaxial anisotropy in the PLD films can then be explained by hard lattice clamping of the Fe lattice to the GaAs(001) which most likely decreases the Fe interface lattice relaxation and its corresponding lattice shear.

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