Ferromagnetic resonance study of Fe superparamagnetic nanoclusters on GaAs(100)

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The fundamental magnetism of the Fe film of 4 ML thick epitaxially grown on GaAs(100) was studied using *ex situ* ferromagnetic resonance (FMR) at room temperature. In parallel geometry the resonance fields were found to be strikingly different along $[0\bar{1}1]$ (2170 Oe) and [011] (3360 Oe) direction, exhibiting a large uniaxial anisotropy with the easy and hard directions along $[0\bar{1}1]$ and [011], respectively. For perpendicular geometry the data of FMR show an abnormally low resonance field of 5050 Oe. It implies a pronounced reduction of effective magnetization. This can be explained by a combination of the shape anisotropy of the nanoclusters with their large perpendicular anisotropy. The *g* factor of these nanoclusters is even smaller than the bulk value. The linewidth is also obviously anisotropic, 220 Oe in easy direction, $[0\bar{1}1]$ and smaller by 50% in hard direction, [011]. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1359458]

I. INTRODUCTION

Ferromagnetic resonance (FMR) is one of the convenient methods for studying magnetic thin films[1] because of its high sensitivity and the possibility of providing information on various important properties, such as the magnetization, magnetic anisotropy, *g* value, exchange constant, by observing the resonance field and spin wave spectra. The linewidth of FMR signal is related to the damping constant of spin dynamics and to the inhomogeneity and defects in the films. In ultrathin films of a few monolayers thick the morphology and microstructure are characteristic of the initial stage of film growth, which causes the magnetic properties different from thick films and bulk material. These features can be observed in FMR measurements.

The previous studies[2] of the epitaxial growth of Fe on GaAs(100) showed that the magnetic properties at room temperature developed via three phases: a nonmagnetic phase for the first 3.5 ML, a short-range-ordered superparamagnetic phase, and a ferromagnetic phase above about 5 ML. Based on the experimental results of low-energy electron diffraction (LEED), *in situ* magneto-optical Kerr effect (MOKE), and *ex situ* alternating field gradient magnetometer (AGFM) measurements it was considered that the film of the average coverage of Fe of 4 ML is composed of superparamagnetic clusters with dimensions about $100\times100\times7.15$ Å,[3] but the magnetization of the film is 1.6×10^3 emu/cm^3, only slightly smaller than the bulk value. In this article we report the results of our further study on the fundamental magnetic properties of a 4 ML film by *ex situ* ferromagnetic resonance.

II. EXPERIMENTS

The epitaxial ultrathin Fe films were grown on GaAs(001) substrate in ultrahigh vacuum with a base pressure of 6×10^{-10} mbar by a multitechnique molecular beam epitaxy system. The preparation of GaAs(001) substrate with a clean and ordered surface for metal film growth was reported elsewhere.[2] Fe was grown at room temperature at a

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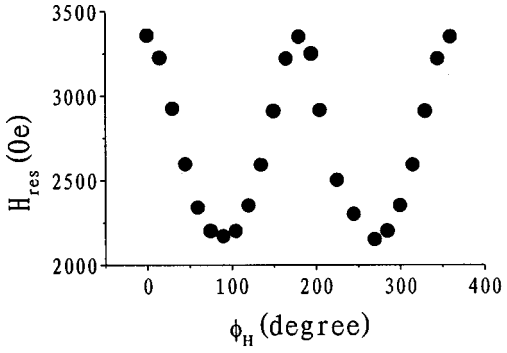

FIG. 1. The experimental data of the field $H_{\rm res}$ as a function of field orientation angle ϕ_H.

rate of about 1 ML/min. The surface structure of the substrate and the Fe film was determined by means of LEED, which showed that Fe grew epitaxially with the epitaxial relationship Fe(001)⟨100⟩‖GaAs(001)⟨100⟩, starting from 5 ML. However, for 4 ML, no Fe LEED spots appeared, demonstrating that the coverage of 4 ML did not form a continuous layer. The *ex situ* ferromagnetic resonance (FMR) measurements were performed with Brucker ESR equipment of model ER-200D-SRC at microwave frequency of 9.78 GHz at room temperature. The maximum sweep field was 14 000 Oe. A small piece of the film of 3×3 mm^2 was mounted at the sample holder, inserted in the microwave cavity, which can have orientations at any angle with respect to the steady field in the film plane and out of plane.

III. EXPERIMENTAL RESULTS AND THEORETICAL DISCUSSION

The experimental data of FMR field $H_{\rm res}$ and linewidth ΔH as a function of field orientation angles, ϕ_H and θ_H, are shown in Figs. 1, 2, and 3. In parallel geometry in Fig. 1 the resonance fields were found to be strikingly different along $[0\bar{1}1]$ (2170 Oe) and [011] (3360 Oe) direction, exhibiting a large uniaxial anisotropy with the easy and hard directions along $[0\bar{1}1]$ and [011], respectively. The corresponding linewidth ΔH is also obviously anisotropic, 220 Oe in easy direction, $[0\bar{1}1]$ and smaller by 50% in hard direction, [011]. For perpendicular geometry the data of FMR show an abnormally low resonance field of 5050 Oe. In order to determine magnetic parameters from the data, let us describe briefly the theory of FMR.

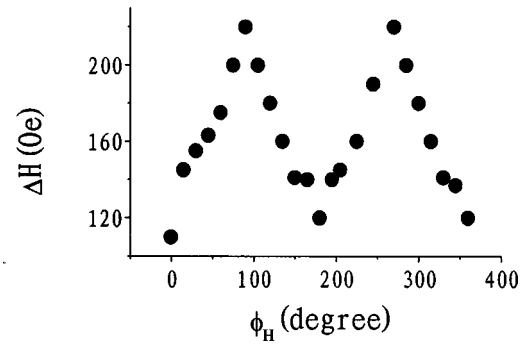

FIG. 2. The experimental data of the linewidth ΔH as a function of field orientation angle ϕ_H.

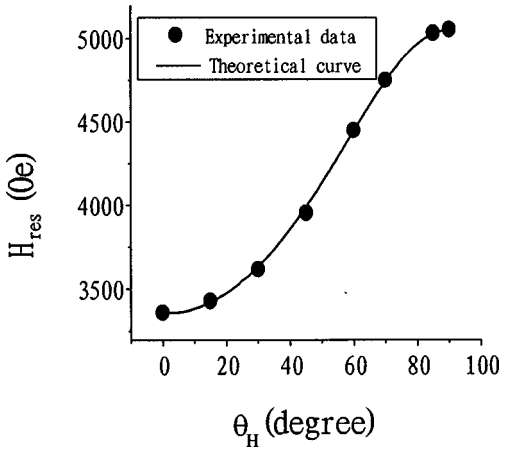

FIG. 3. The theoretical and experimental data of resonance field as a function of θ_H.

The well-known Landau–Lifshitz–Gilbert equation of the precession of the magnetization as follows is used for describing dynamics of FMR[1,3]

$$\frac{d\mathbf{M}}{dt}=-\gamma\mathbf{M}\times\mathbf{H}+\frac{\eta}{\gamma M^2}\left[\mathbf{M}\times\frac{d}{dt}\mathbf{M}\right]. \tag{1}$$

The first term refers to the torque produced by the effective field, which may include the external steady field H, microwave field h, and various effective fields originated from magnetic anisotropies. The second is the Gilbert damping term. The term γ is the gyromagnetic ratio, $\gamma=ge/2mc$ and g is the Landé splitting factor. For free electrons, $g=2.0023$, and $\gamma=1.76\times10^7$/s Oe. For bulk Fe a reasonable value of $g=2.134$ was reported.[3] η is the Gilbert damping constant.

A simple way to determine the resonance frequency is given by considering the equilibrium condition of magnetization under steady field and neglecting the dynamical aspects of FMR and damping effect, leading to the following general expression of resonance frequency:[4]

$$\left(\frac{\omega}{\gamma}\right)^2=\frac{1}{(M\sin\theta)^2}\{F_{\theta\theta}F_{\phi\phi}-(F_{\theta\phi})^2\}. \tag{2}$$

Here F_{ij} is the second derivative of the free energy density F with respect to the spherical coordinates θ and ϕ for the single crystalline thin film system. For the data in the figures, F is assumed to be as follows:

$$F=-MH[\cos\theta\cos\theta_H+\sin\theta\sin\theta_H\cos(\phi-\phi_H)]$$
$$-2\pi M^2\sin^2\theta+K_u^{\perp}\sin^2\theta-k_1^{\parallel}\sin^2\theta\sin^2\phi \tag{3}$$

which contains Zeeman energy, demagnetzing energy, perpendicular anisotropy energy, and in-plane uniaxial anisotropy energy. Substituting Eq. (3) into Eq. (2) and considering the equilibrium condition of magnetization orientation by minimizing the free energy with respecting to θ and ϕ, the following expression for resonance frequency as a function of field orientation, θ_H and ϕ_H, is obtained:

$$\left(\frac{\omega}{\gamma}\right)^2 = \{H[\cos\theta\cos\theta_H + \sin\theta\sin\theta_H\cos(\phi-\phi_H)] - 4\pi M_{\text{eff}}\cos 2\theta + H_K^{\parallel}\cos 2\theta\sin^2\phi\} \times \{H[\cos\theta\cos\theta_H + \sin\theta\sin\theta_H\cos(\phi-\phi_H)] - 4\pi M_{\text{eff}}\cos^2\theta + H_K^{\parallel}(\cos^2\phi - \sin^2\theta\sin^2\phi)\} - \{H_K^{\parallel}\cos\theta\cos\phi\sin\phi\}^2. \quad (4)$$

Here $4\pi M_{\text{eff}} = 4\pi M - 2K_u^{\perp}/M$. $4\pi M$ refers to the demagnetizing field and $2K_u^{\perp}/M$ is the effective perpendicular anisotropy field, $H_K^{\perp}$, and $H_K^{\parallel} = 2K_1^{\parallel}/M$ is the effective in-plane uniaxial anisotropy field. By theoretical fitting of Eq. (4) with the experimental data of resonance field at different ϕ_H and θ_H, the magnetic parameters $4\pi M_{\text{eff}}$, $H_K^{\parallel}$, and γ or g factor are determined. Figure 3 also shows the theoretical curves of Eq. (4) with the following parameters $4\pi M_{\text{eff}} = 1900\,\text{G s}$, $\gamma = 1.78\times 10^7/\text{s Oe}$, $g = 2.02$ and $H_K^{\parallel} = 740\,\text{Oe}$ when $\phi_H = \phi = \pi/2$.

A discussion for the figures and the fitted data is given as follows.

(1) The value of $\pi M_{\text{eff}} = 1900\,\text{G s}$ is much smaller than the value found previously by AGFM, where $4\pi M = 4\pi \times 1600\,\text{G s}^2$. It implies that there is a strong perpendicular anisotropy in addition to the shape anisotropy of the nanoclusters which is smaller than a continuous plane. From the estimated dimensions of the nanoclusters[2] we assume that the shape anisotropy field in Eq. (4) is not $4\pi M$, but $(N_{\perp} - N_{\parallel})M \sim 10.7\times M = 16\,400\,\text{G s}$. The perpendicular anisotropy constant is then determined to be $K_u^{\perp} = 1.16\times 10^7\,\text{erg/cm}^3$. Considering the average thickness of the film, $4\,\text{ML} = 5.7\,\text{Å}$, the equivalent surface anisotropy constant $K_S = 0.6\,\text{erg/cm}^2$. A rather large value.

(2) The value of $g = 2.02$ is even smaller than the value of bulk Fe. It is puzzling since for the ultrathin films more contribution of electron orbital moment is expected to appear to the magnetization.

(3) In-plane anisotropy becomes uniaxial instead of that of fourfold symmetry in cubic structure. This is consistent with the *in situ* MOKE measurement.[2] The effective in-plane anisotropy field $H_K^{\parallel} = 740\,\text{Oe}$ refers to $K_1^{\parallel} = 6\times 10^5\,\text{erg/cm}^3$. Also a relatively large value.

(4) The linewidth is relatively large and anisotropic, 220 Oe in easy direction, $[0\bar{1}1]$, while 110 Oe in hard direction, [011]. It is consistent with severe inhomogeneity in the 4 ML coverage of Fe and the in-plane uniaxial anisotropy of the clusters.

ACKNOWLEDGMENTS

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