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Temperature dependences of surface magnetoelastic constants of ultrathin Fe/GaAs (001) films

R. Żuberek, K. Nesteruk, K. Fronc, S. Piechota, and H. Szymczak

Institute of Physics, Polish Academy of Sciences, 32/46 Al. Lotników, 02-668 Warsaw, Poland
E-mail: szymh@ifpan.edu.pl

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The magnetoelastic constants of epitaxial iron films prepared by dc magnetron sputtering on single crystal GaAs (001) substrate in argon atmosphere and covered with a protective Si layer have been investigated in the temperature range 10–300 K by means of the strain modulated ferromagnetic resonance. It has been shown that the magnetoelastic constants strongly depend on the thickness of the film. The surface components of the magnetoelastic constants have been determined and analyzed within the Néel and dipolar models. The proposed analysis of experimental data gives chance for deeper insight into mechanisms responsible for magnetostriction of iron thin films.

PACS: 75.70.Cn Magnetic properties of interfaces (multilayers, superlattices, heterostructures);
75.80.+q Magnetomechanical effects, magnetostriction;
75.70.Ak Magnetic properties of monolayers and thin films.

Keywords: magnetoelastic constants, ferromagnetic resonance, iron films.

1. Introduction

The films of iron deposited on GaAs have been of strong interest for their possible applications in magnetoelectronic devices. It was shown that Fe films with thickness of 5 monolayers or more epitaxially grown at room temperature (RT) on GaAs (001) surfaces, are ferromagnetically ordered at room temperature with nearly bulk magnetic moment per atom [1]. It is generally accepted that magnetic anisotropy essentially control the hysteretic behavior of ferromagnets and consequently determine most of the parameters (e.g., coercivity, permeability, energy of magnetic domain walls) important for practical applications. Therefore, the understanding of magnetic anisotropies in Fe/GaAs films is of crucial importance for the development of various spintronic devices. In this particular case the magnetic anisotropy of the thin films is determined to a large degree by surface or interface effects. The interfacial anisotropy does exist not only in the out-of-plane direction, but can also arise within the plane of the film. In thin single crystal Fe films on GaAs (001) substrate an uniaxial anisotropy is observed with easy axis in the [110] direction [2] below a critical film thickness. The uniaxial magnetic anisotropy has already been observed in thicker Fe films on GaAs (001) [3,4]. There have been speculations on the origin of this anisotropy. It was suggested that this anisotropy is related to the presence of a $Fe_3Ga_{2-x}As_x$ at the interface [4]. More results, however, support the explanation of the uniaxial term by the intrinsic anisotropy of the dangling bonds at the GaAs (001) surface [2,3,5,6].

The thickness and stress dependence of magnetoelastic constants of iron films have also been reported by several authors (see, e.g., [5,7]). In the present paper we continue our studies of the magnetoelastic constants of the iron films with different thickness but with similar stresses at the surface/interface [6,8]. To explore this problem further the epitaxial iron films prepared by dc magnetron sputtering on single crystal GaAs (001) substrate in argon atmosphere and covered with a protective Si layer have been investigated from 10 to 300 K by means of the strain modulated ferromagnetic resonance (SMFMR).

2. Experimental

The Fe films in form of a wedge and the thickness range from 3 to 6 nm, were grown by dc magnetron sputtering on GaAs (001) single crystal substrate at room temperature. Ar were used as a sputter gas at pressures of $3 \cdot 10^{-1}$ Pa. Prior to the thin film deposition, polished semiinsulating GaAs (001) substrates were cleaned in trichloroethylene, methanol and rinsed in deionized water. Then the substrates were dipped into a solution of $6H_2SO_4$:$1H_2O_2$:$1H_2O$ (by volume) for 15 min and rinsed in deionized water. Next the GaAs substrates were dipped into solution of $10H_2O{:}1HCl$ without subsequent rinsing with ultrapure water. After etching procedure the substrate was immediately mounted to the substrate holder and loaded into loadlock chamber of sputtering system. The time needed to transfer the substrate from HCl solution into UHV environment with pressure 10^{-6} Pa not exceeded 20 min. In order to prevent oxidation of magnetic layer, the iron films were covered with a protective Si layer of 7.5 nm thick.

3. Ferromagnetic resonance conditions

SMFMR experiments were performed using a standard X-band spectrometer operating at 9.2 GHz with 100 kHz field modulation and 130 kHz strain modulation. The dc magnetic field was applied in the plane of the film along the [100] and [110] directions. The SMFMR spectra were analyzed using a coordinate system in which the magnetization of the film **M** and the external magnetic field **H** make angles θ and θ_H with respect to the film normal and angles ϕ and ϕ_H with respect to the [100] axis of the film. In our experiment $\theta_H = \pi/2$.

In order to calculate the resonance frequency we have used the approach developed by Suhl [9]. Considering the uniaxial and cubic anisotropy energies the free energy density of the system is

$$F = -MH \sin\theta \cos(\phi - \phi_H) + 2\pi M_{\mathrm{eff}}^2 \cos^2\theta + \\ + \frac{K_1}{4}(\sin^4\theta \sin^2 2\phi + \sin^2 2\theta) - K_u \sin^2\theta \cos^2(\phi - 45^\circ) . \tag{1}$$

It is useful to define the anisotropy fields

$$H_k = 2K_1/M_s , \quad H_u = 2K_u/M_s . \tag{2}$$

The terms containing K_u represents in plane uniaxial anisotropy energy, K_1 is cubic anisotropy constant. The effective magnetization term consists of saturation magnetization and out of plane anisotropy field:

$$4\pi M_{\mathrm{eff}} = 4\pi M_s + 2K^{\perp}/M_s . \tag{3}$$

The equilibrium conditions for the magnetization and the resonance condition for the FMR can be found using the following equations [9]:

$$\frac{\partial F}{\partial\theta} = \frac{\partial F}{\partial\phi} = 0, \quad \left(\frac{\omega}{\gamma}\right)^2 = \frac{1}{M_s^2 \sin^2\theta}\left[\frac{\partial^2 F}{\partial\theta^2}\frac{\partial^2 F}{\partial\phi^2} - \left(\frac{\partial^2 F}{\partial\theta\partial\phi}\right)^2\right], \tag{4}$$

where ω is the circular frequency, γ is the giromagnetic ratio.

For the strain modulated FMR (SMFMR) the magnetoelastic energy [10] should be added to the free energy of the system:

$$F_{ME} = B_{ijkl}\alpha_i\alpha_j\varepsilon_{kl} + D_{ijklmn}\alpha_i\alpha_k\varepsilon_{kl}\varepsilon_{mn} = B_{ijkl}^{\mathrm{eff}}\alpha_i\alpha_j\varepsilon_{kl} , \tag{5}$$

where the first term is linear and second nonlinear functions of the strain. For cubic crystal the linear part of the magnetoelastic energy can be written as

$$F_{ME} = b_1(\alpha_1^2\varepsilon_{11} + \alpha_2^2\varepsilon_{22} + \alpha_3^2\varepsilon_{33}) + \\ + 2b_2(\alpha_1\alpha_2\varepsilon_{12} + \alpha_2\alpha_3\varepsilon_{23} + \alpha_1\alpha_3\varepsilon_{13}) , \tag{6}$$

$$b_1 = B_{1111} - B_{1122}, \quad b_2 = 2B_{1313} , \tag{7}$$

where b_1 and b_2 are the magnetoelastic constants, the ε_{ij} and α_i are the components of strain tensor and the direction cosines of **M** with respect to the cubic axes, respectively.

For thin films, in which the thickness (t) of the film is smaller than the exchange length, the effective anisotropy and magnetoelastic constants can be written as ([11] and reference therein)

$$K_{(n)}^{\mathrm{eff}} = K_{(n)}^{v} + \frac{2}{t} K_{(n)}^{s} , \tag{8}$$

where

$$2K_{(n)}^{s} = K_{(n)}^{s1} + K_{(n)}^{s2} \tag{9}$$

and

$$b_{(n)}^{\mathrm{eff}} = b_{(n)}^{v} + \frac{2}{t} b_{(n)}^{s} , \tag{10}$$

$n = u$ for uniaxial and 1 for in-plane cubic anisotropy and $n = 1$ or 2 for magnetoelastic constants. In (9) the contributions from both interfaces are considered.

4. Experimental results

The SMFMR measurements with dc magnetic field parallel to [110] direction allow determination of the effective magnetoelastic constant b_2^{eff}. As examples the FMR derivative spectra for the film with 4 nm thickness for magnetic and strain modulation measured at 10 K and RT are shown in Fig. 1. The temperature dependence of b_2^{eff}

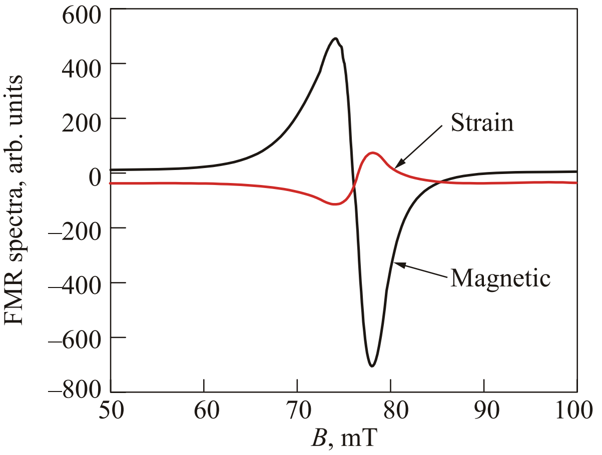

Fig. 1. The typical FMR spectra for field directed along [110] axis for magnetic and strain modulation measured at 10 K and RT for the Fe film with the thickness of 4 nm.

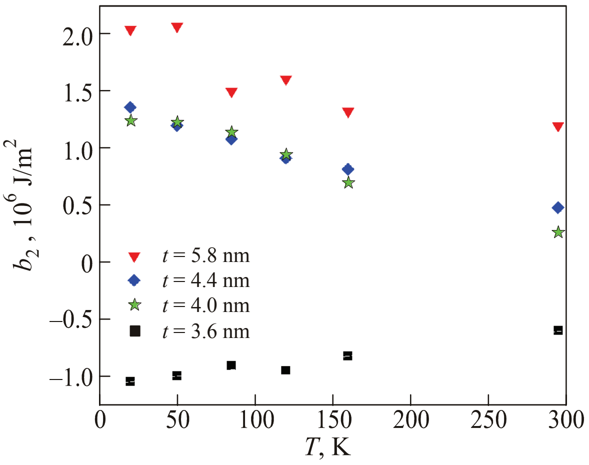

Fig. 2. The temperature dependence of b_2^{eff} for films with different thicknesses.

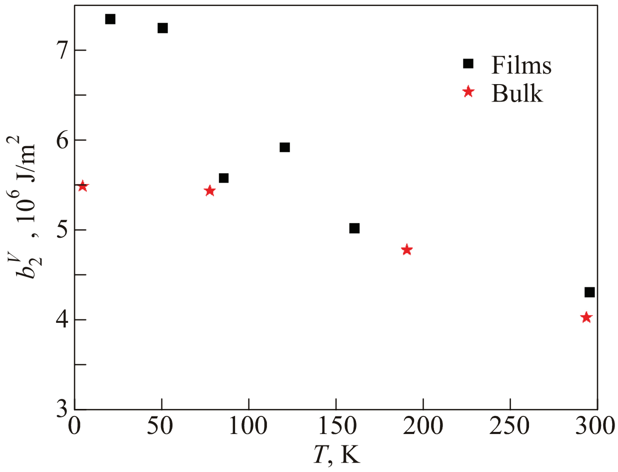

Fig. 4. The fitted values of the volume magnetoelastic constants for several temperatures. The values of magnetoelastic constant b_2 of bulk bcc Fe are also displayed.

for the films with different thicknesses are shown in Fig. 2. The fitted values of the volume and surface (interface between GaAs and Fe from one side and Fe/Si from the second one) magnetoelastic constants for several temperatures are shown in Figs. 3 and 4. The values of the magnetoelastic constant b_2 of bulk bcc Fe [12,13] are also displayed. The surface magnetoelastic constants seem to have origin similar to that established for the surface anisotropy.

5. Comparison with the Néel and dipolar models

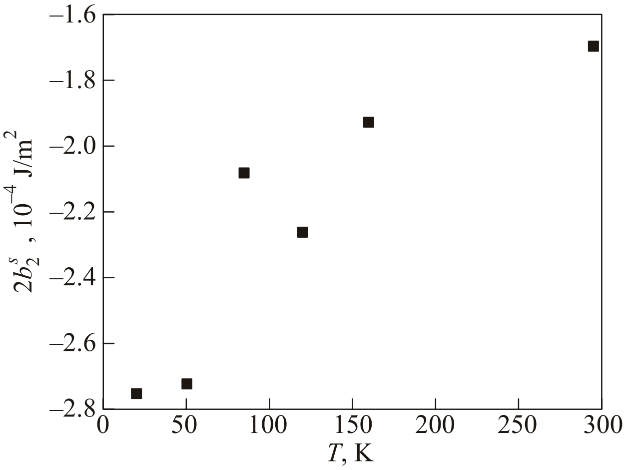

Fig. 3. The fitted values of the surface (interface between GaAs and Fe from one side and Fe/Si from the second one) magnetoelastic constant b_2 for several temperatures.

Usually, in order to compare the surface in-plane cubic and uniaxial anisotropy constants with the Néel [14,15] or dipolar [16,17] models the effective anisotropy constant is written in another way [18]. Assuming that the thickness of one monolayer is equal d then the thickness of the film will be equal $t = Nd$ where N is the number of atomic layers in the film. In ideal structure of the film there are two surface layers of thickness d and $N-2$ volume layers. Then effective anisotropy constant can be written as

$$K^{\text{eff}}_{(n)} Nd = K^{v}_{(n)}(N-2)d + 2K^{ss}_{(n)}, \qquad (11)$$

where

$$2K^{ss}_{(n)} = K^{ss1}_{(n)} + K^{ss2}_{(n)}, \qquad (12)$$

and $K^{ss}_{(n)}d$ is the proper surface anisotropy constant calculated, e.g., in the Néel or dipolar model. The following dependence between $K^{s}_{(n)}$ and $K^{ss}_{(n)}$ results from Eqs. (1) and (4):

$$K^{s}_{(n)} = K^{ss}_{(n)} - K^{v}_{(n)}d\,. \qquad (13)$$

It means that the measured surface anisotropy constant ($K^{s}_{(n)}$) depends additionally on volume contribution (K_1^v).

In some cases [11] the volume anisotropy constant for the uniaxial anisotropy is equal to zero and therefore both surface anisotropy constants are the same ($K^{s}_{(u)} = K^{ss}_{(u)}$). The in-plane surface cubic anisotropy constant obtained from linear dependence of the effective anisotropy constant on inverse film thickness is less than calculated one by dK_1^v. For the films with the same surface anisotropy constant but different volume anisotropy constant the linear dependence surface anisotropy constant (K_1^s) on the volume anisotropy constant (K_1^v) is observed (see for instance Fig. 4 in paper [11]).

Usually magnetic anisotropy and magnetostriction have the same origin. Therefore, in the case of magnetoelastic constant the same problem appears as in magnetic anisotropy constant and, consequently, the same formula for simulating surface magnetoelastic constant (b_2^s) and proper surface magnetoelastic constant (b_2^{ss}) should be used

$$b_2^s = b_2^{ss} - b_2^v d\,. \qquad (14)$$

According to literature (e.g., [19]) the dependence of the magnetoelastic constants on the thickness of magnetic layer arises as an intrinsic or an extrinsic effect. The intrinsic effect is related to the broken symmetry of atoms at the interface (see, e.g., the well-known Néel model [14,15]).

According to the Néel model the surface magnetoelastic tensor, B_{ijkl}^{ss}, for the bcc structure and for (001) surface has cubic symmetry and magnetoelastic constant (b_2^{ss}) is equal to [15]

$$b_2^{ss} = 2B_{1313}^{ss} = \frac{4}{9} n_s (p + mr), \qquad (15)$$

where n_s is the density of surface atoms and p for the bcc iron is equal to $-1.5326 \cdot 10^{-23}$ J, $mr = 12.0 \cdot 10^{-23}$ J. Then

$$b_2^{ss} = 0.57 \cdot 10^{-3} \text{J/m}^2, \text{ and } b_2^{s} = -0.5 \cdot 10^{-3} \text{ J/m}^2 \qquad (16)$$

which is about several times smaller than values obtained in experiment.

The extrinsic effects arise mainly due to the misfit dislocations, interdiffusion and even due to the surface roughness. The relatively large scatter of the experimental data seen in Figs. 3, 4 suggests that interface Fe–GaAs is not flat but rather rough. The roughness was shown to give considerable contribution to the surface anisotropy and magnetostriction [20]. The roughness depends on various factors which are difficult to remove.

5. Conclusion

We have grown epitaxial iron films on single crystalline GaAs (001) substrates with Ar as a sputtering gas. The films were covered with Si overlayers. The magnetostriction constants have been measured by SMFMR method. The small FMR linewidth of the measured films indicates a high crystalline quality of the Fe layers. It has been found that magnetostriction constants are composed of surface and bulk contributions dependend on temperature. The bulk magnetoelastic constants were equal to the values found for bulk Fe. The surface anisotropy and surface magnetostriction are related first of all to the effects of broken symmetry of atoms at the interfaces. This observation seems to be related to tetragonal strain due to lattice mismatch.

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