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FMR Study of the Anisotropic Properties of an Epitaxial Fe_3Si Film on a Si(111) Vicinal Surface

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The anisotropic characteristics of an iron silicide (Fe_3Si) epitaxial thin magnetic film grown on a Si(111) silicon vicinal surface with a misorientation angle of 0.14° have been measured by the ferromagnetic resonance method. It has been shown that the polar and azimuth misorientation angles of the crystallographic plane of the substrate can be determined simultaneously from the angular dependences of the ferromagnetic resonance field of the epitaxial film. The effective saturation magnetization of the film $M_{\text{eff}} = 1105$ G and the constant of the cubic magnetocrystalline anisotropy $K_4 = 1.15 \times 10^5$ erg/cm^3 have been determined. The misorientation of the substrate plane leads to the formation of steps on the film surface and, as a result, to the appearance of uniaxial magnetic anisotropy of the magnetic dipole nature with the constant $K_2 =$ 796 erg/cm^3. Small unidirectional magnetic anisotropy ($K_1 = 163$ erg/cm^3), which may be associated with symmetry breaking on the steps of the film and is due to the Dzyaloshinskii–Moriya interaction, has been detected.

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1. Epitaxial ferromagnetic films and different layered structures grown on single-crystal substrates are studied actively owing to their usage as elements of magnetoelectronic devices [1, 2]. Traditional semiconductor technologies used in microelectronics are, as a rule, applied for the preparation of such structures. Particular attention has recently been focused on the study of magnetic films grown on the vicinal cuts of Si(111) single-crystal substrates, for which methods of creating stepwise surfaces providing a high accuracy of the step width and step height values are well developed [3, 4]. This provides the possibility of controlling the magnetic properties of the films in a wide range by varying the misorientation angle of the Si(111) vicinal surface in a narrow range [5, 6]. In particular, experiments show that considerable changes in the anisotropic properties of thin magnetic films are observed even when the angles of deviation of the substrate surface from the singular plane are less than one degree [7]. However, the nature of the magnetic anisotropy of films on the vicinal surfaces is not completely clear. This problem was studied in many works [5–8], where different methods were used to quantitatively determine the magnetic anisotropy, including the ferromagnetic resonance (FMR) method having not only sufficient sensitivity but also high accuracy of the measurement [9].

In this work, the magnetic properties of the epitaxial Fe_3Si thin film grown on the Si(111) silicon vicinal surface with a small (on the order of 0.1°) misorientation angle are studied by the FMR method in order to investigate the nature of the observed features of the magnetic anisotropy in such samples.

2. The Fe_3Si thin film was prepared by the method of the simultaneous thermal evaporation of iron and silicon from two crucibles in superhigh vacuum (1.3×10^{-8} Pa) with the subsequent deposition of atoms on the Si(111) boron-doped atomically pure vicinal substrate (with a misorientation angle of about 0.1°) as described in [10]. The epitaxial growth was controlled in situ by a high-speed laser ellipsometer and by the reflected fast electron diffraction method. The structure and phase composition of the sample were determined by transmission electron microscopy and X-ray structural analysis methods. It was established that the thin film was Fe_3Si single-crystal silicide with the unit cell parameter $a = 0.564$ nm and the interplane distance $h = 0.33$ nm. The measured thickness d of the magnetic film was 20 nm.

Magnetic properties on local regions of the Fe_3Si film were studied on an automated scanning ferromagnetic resonance spectrometer [11], which made it possible to record FMR spectra at different directions

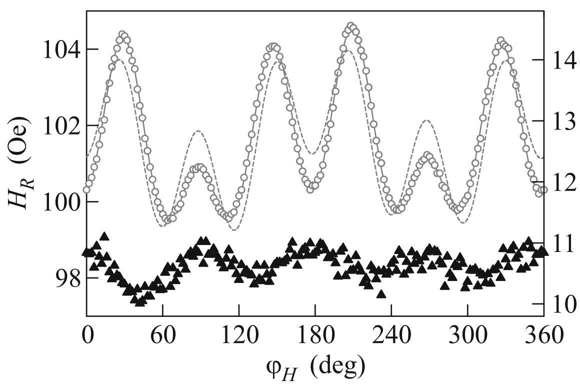

Fig. 1. Resonance field H_R and the FMR linewidth ΔH versus the direction of the sweeping magnetic field φ_H: experimental points in comparison with the calculations for the model of the film on (solid line) the vicinal surface with $\delta = 0.14°$ and (dashed line) the singular surface with $\delta = 0°$.

of the sweeping magnetic field. A miniature microstrip resonator fabricated on the substrate with a high relative permittivity was used in it as a microwave probe. Near the high-frequency magnetic field antinode, a measuring hole with a diameter of 1 mm was made in the resonator shield, which provided the locality of the measurements. The resonance absorption of the microwave power by the studied area of the film sample was as usual recorded in the spectrometer by the modulation method [12] from the variation of the Q-factor of the resonator at the variation of the static magnetic field applied in the film plane. The main advantage of the scanning spectrometer is its high sensitivity resulting from the large filling coefficient of the microstrip resonator by the measured sample area owing to the miniature resonator.

Figure 1 shows the dependences of (open circles) the resonance field H_R and (closed triangles) the FMR linewidth ΔH on the direction of the external sweeping magnetic field φ_H taken with a step of 2° at the pump frequency f_0 = 3.329 GHz. In the experiment, the sweeping field and the linearly polarized high-frequency magnetic field were oriented in the film plane and their directions were mutually orthogonal. It can be seen that the studied sample has a complicated angular dependence $H_R(\varphi_H)$, in which six maxima and six minima are pronounced. The analysis of the measured dependence makes it possible to assume that the uniaxial magnetic anisotropy (of the second order) and, possibly, the unidirectional anisotropy (of the first order) are present in the studied Fe_3Si film along with the cubic anisotropy (fourth-order anisotropy). It is also seen that the studied sample has a relatively narrow line of uniform FMR, the width of which varies within one oersted (10–11 Oe). The dependence $\Delta H(\varphi_H)$ shows only four minima and four maxima as it takes place in films with uniaxial magnetic anisotropy [13].

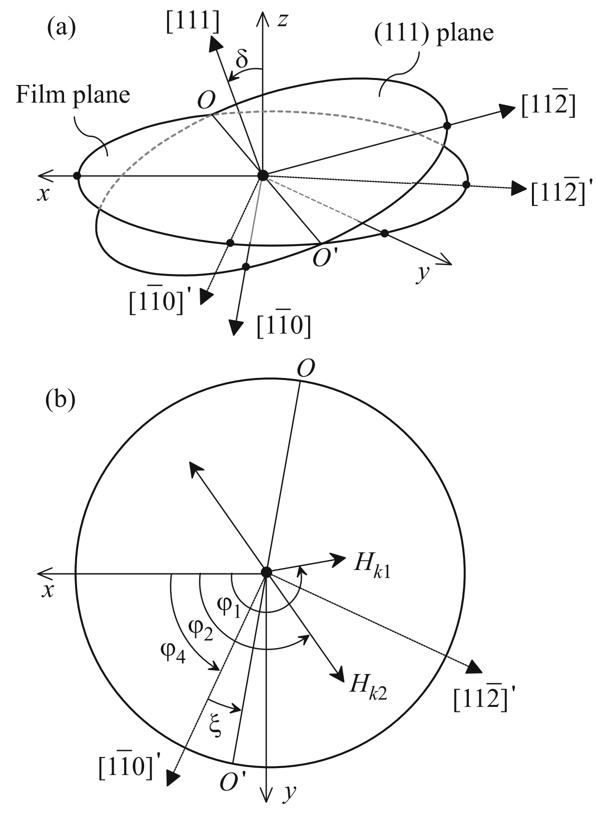

Fig. 2. (a) Coordinate system and scheme of the location of the (111) crystallographic plane of the Fe_3Si single-crystal film with respect to its surface. (b) Notation of the phenomenological model of the thin film (top view).

3. To analyze and interpret experimental results, we consider the model of the studied iron silicide film deposited on the (111) vicinal surface with the small misorientation angle δ (Fig. 2a) which is measured from the z axis orthogonal to the film surface in the Cartesian coordinate system. Figure 2b shows the film surface in which the directions of the anisotropy fields (easy magnetization axes) resulting from the unidirectional anisotropy H_{k1} and uniaxial anisotropy H_{k2} are shown. Here, φ_1, φ_2, and φ_4 are the angles of the direction of anisotropy fields and the $[1\bar{1}0]'$ crystallographic direction, respectively, measured from the x axis.

Using a phenomenological approach, we theoretically analyze the model of an infinite uniformly magnetized thin magnetic film. The saturation magnetization of the film is M_s and the magnetization vector **M** in the Cartesian coordinate system xyz (Fig. 2) is characterized by the polar angle θ formed with the film plane and the azimuth φ measured from the x axis. Let the static magnetic field **H** be oriented in the film plane at the angle φ_H also measured from the x axis. In this case, the free energy density can be presented as the sum

$$
\begin{aligned}
F(\theta,\varphi) = &-M_s H\cos(\varphi-\varphi_H)\sin\theta \\
&+ 2\pi M_s^2\cos^2\theta - K_\perp\cos^2\theta \\
&- K_1\sin\theta\cos(\varphi-\varphi_1) \\
&- K_2\sin^2\theta\cos^2(\varphi-\varphi_2) + F_{mc}.
\end{aligned} \tag{1}
$$

Here, the first and second terms describe the contributions of the Zeeman energy and the energy of the demagnetizing field of the film, respectively. The third term describes the energy of the uniaxial orthogonal anisotropy with the constant $K_\perp$, which, as is known, takes into account the symmetry breaking on the surface of a thin film and at the interface between the film and substrate. The next two terms in Eq. (1) describe the energies of the unidirectional magnetic anisotropy with the constant K_1 and uniaxial magnetic anisotropy with the constant K_2 whose fields H_{k1} and H_{k2} are in the film plane and are directed at the angles φ_1 and φ_2, respectively (Fig. 2b). The last term F_{mc} is the energy of the magnetocrystalline anisotropy associated with the magnetic anisotropy of the cubic crystal.

It is known that the energy of the magnetic anisotropy with the constant K_4 for crystals of the cubic system is written as $F_{mc} = K_4(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_1^2\alpha_3^2)$, where α_1, α_2, and α_3 are the direction cosines of the magnetization vector **M** with respect to the [100], [010], and [001] fourth-order axes, respectively. In the case where the single-crystal thin films are formed on the (111) vicinal surfaces, which are characterized by small misorientation angles δ usually less than 5°, it is possible to use the linear approximation of the expansion of the energy in the small parameter δ in the calculation of F_{mc}. Then, for the geometry of a thin film shown in Fig. 2, the magnetocrystalline anisotropy has the form

$$
\begin{aligned}
F_{mc} = &K_4\left(\frac{1}{3}\cos^4\theta + \frac{1}{4}\sin^4\theta\right) \\
&- \frac{K_4\sqrt{2}}{3}\sin^3\theta\cos\theta\sin^3(\varphi-\varphi_4) \\
&+ \delta\frac{K_4}{6}\{\sin 2\theta\times(7\sin^2\theta-4)\sin(\varphi-\varphi_4+\xi) \\
&+ \sqrt{2}\sin^2\theta\times(7\sin^2\theta-6)\cos 2(\varphi-\varphi_4-\xi/2) \\
&- \sqrt{2}\sin^4\theta\times\cos 4(\varphi-\varphi_4+\xi/4)\}.
\end{aligned} \tag{2}
$$

It is important to note that the contribution of the energy of the demagnetizing field owing to the shape anisotropy in low-dimension systems such as thin films usually dominates in the total energy of the magnetic anisotropy. Therefore, the shape anisotropy is mainly responsible for the planar orientation of the magnetization in the sample. However, we note that the relative contribution of the energy of the surface anisotropy for ultrathin films whose thickness does not exceed several atomic monolayers becomes considerable; consequently, the orientation of the spontaneous magnetization normal to the film plane is possible [14].

To calculate the frequency of the uniform ferromagnetic resonance ω_0, it is convenient to use the Smit–Suhl formula [15, 16], which requires the calculation of double partial derivatives $F_{\theta\theta}$, $F_{\varphi\varphi}$, and $F_{\theta\varphi}$ of the expression for the free energy distribution density over the angles of the orientation of the magnetization θ and φ in the spherical coordinate system:

$$
\omega_0 = \frac{\gamma}{M_s\sin\theta_M}[F_{\varphi\varphi}F_{\theta\theta} - F_{\theta\varphi}]^{1/2}_{\theta_M,\varphi_M}, \tag{3}
$$

where γ is the gyromagnetic ratio. The partial derivatives are calculated for the angles $\theta = \theta_M$ and $\varphi = \varphi_M$ corresponding to the equilibrium position of the magnetization vector **M**, which obviously corresponds to the minimum of the free energy F. Therefore, the angles θ_M and φ_M are determined from the equations

$$
F_\varphi(\theta_M,\varphi_M) = 0, \quad F_\theta(\theta_M,\varphi_M) = 0. \tag{4}
$$

When the ferromagnetic resonance spectra are recorded under the sweep of the static magnetic field H, the resonance field H_R for any fixed frequency $\omega_0 = 2\pi f_0$, where f_0 is the microwave pump frequency, which, as was already noted, was 3.329 GHz in the experiment, can be easily calculated from Eqs. (3) and (4).

4. It is obvious that, for the agreement of the theoretical dependence of the FMR field calculated under the variation of the direction of the static sweeping magnetic field according to Eqs. (3) and (4) with the experimental dependence $H_R(\varphi_H)$ (see Fig. 1), it is necessary to choose all parameters of the phenomenological model of the studied film. For convenience and in order to accelerate the selection of the model parameters, an iteration procedure of the numerical analysis was elaborated making it possible to automate the process of the approximation of the experimental dependence $H_R(\varphi_H)$ by the corresponding theoretical dependence in order to minimize the divergence between theory and experiment.

If it is assumed in the theoretical model of the film that the misorientation angle of the vicinal surface is $\delta = 0°$; i.e., the experimental sample is grown on the Si(111) singular surface, then the theoretical dependence $H_R(\varphi_H)$ obtained by the approximation of the experimental points (dashed line in Fig. 1) will reproduce the experiment only qualitatively. The optimal parameters of the phenomenological model of the Fe_3Si film providing the minimum deviation between theory and experiment for this case are presented in the first line in the table. Here, the following notation is used: $M_{eff} = M_s - K_\perp/2\pi M_s$ is the effective saturation magnetization, $H_{k1} = K_1/M_{eff}$ is the unidirectional anisotropy field, $H_{k2} = 2K_2/M_{eff}$ is the uniaxial anisot-

Parameters of the phenomenological model of the thin Fe_3Si film obtained by the approximation of the experimental dependence $H_R(\varphi_H)$

Substrate Si(111)	M_{eff}, G	K_1, erg/cm^3 (H_{k1}, Oe)	φ_1, deg	K_2, erg/cm^3 (H_{k2}, Oe)	φ_2, deg	K_4, erg/cm^3 (H_{k4}, Oe)	φ_4, deg	ξ, deg	δ, deg
Singular	1105.4	169 (0.15)	64.66	644 (1.17)	90.30	1.152×10^5 (208.34)	87.96	0	0
Vicinal	1105.3	163 (0.15)	64.81	796 (1.44)	90.38	1.149×10^5 (207.93)	87.94	5.00	0.14

ropy field, and $H_{k4} = 2K_4/M_{eff}$ is the cubic anisotropy field.

The solid line in Fig. 1 shows the theoretical dependence $H_R(\varphi_H)$ plotted for the film model corresponding to the experimental sample grown on the vicinal surface of the substrate ($\delta \neq 0°$). In this case, the approximation of the experimental points providing the minimum deviation between theory and experiment was performed with the variation of the magnetic parameters of the phenomenological model of the studied Fe_3Si film, as well as the angles δ and ξ of the misorientation of its surface (see Fig. 2). The optimal parameters obtained in this manner are presented in the second line of the table. It can be seen that the iron silicide film prepared in the Si(111) vicinal surface is misoriented in two directions: the polar angle of the (111) facet is deviated by $\delta = 0.14°$ in the $[11\bar{2}]$ direction and the azimuth is deviated by $\xi = 5°$ in the $[\bar{1}10]$ direction (see Fig. 2).

It was shown in [17] that the contribution of the orthogonal surface anisotropy $K_\perp$ to the effective saturation magnetization becomes insignificant for Fe_3Si films thicker than 20 atomic monolayers. Since the thickness of the studied Fe_3Si film is on the order of 60 monolayers, it can be accepted that the saturation magnetization of the sample is $M_s \approx M_{eff} = 1105.3$ G. It is important to note that the M_s value obtained is about 10% higher than that for the Fe_3Si film [18] grown on Si(111) ($M_s \approx 1000$ G) and nearly 14% higher than that for the Fe_3Si film [17] grown on GaAs(001) ($M_s \approx 970$ G). However, this magnetization is about 9% lower than that of bulk Fe_3Si ($M_s \approx 1248$ G) [19]. The value obtained for the magnetocrystalline anisotropy constant $K_4 = 1.149 \times 10^5$ erg/cm^3 is in good agreement with the value for bulk Fe_3Si (~1.08×10^5 erg/cm^3) [19] but is about twice as high as that for Fe_3Si grown on GaAs(001) (~6×10^4 erg/cm^3) in [17].

It is known that additional uniaxial magnetic anisotropy appears in thin ferromagnetic films formed on the vicinal surface [8]. The nature of this anisotropy can be associated with the symmetry breaking on the stepwise surface of the film [20] and with the elastic stresses arising at the interface between the film and substrate owing to the mismatch between the lattice parameters [21]. In addition, the important source of the uniaxial magnetic anisotropy is the long-range magnetic dipole interaction, which makes the magnetization direction along the steps more favorable owing to the magnetic charges induced on the steps [22].

To estimate the degree of the effect of the demagnetizing fields arising because of the presence of steps on the surface and interface of the Fe_3Si/Si(111) structure, we used the formula $N \approx \pi\langle p^2\rangle/\lambda d$ for the demagnetization factor proposed by Schlömann [23]. Here, $\langle p^2\rangle$ is the mean square deviation of the film surface from the average value and λ is the spatial period of these deviations. For the sample geometry used in our work, the magnetic dipole anisotropy field is

$$H_u \approx 4\pi M_s \frac{2\pi}{3}\frac{h}{d}\tan\delta. \quad (5)$$

As a result, the uniaxial magnetic dipole anisotropy field is $H_u = 1.21$ Oe, which is in very good agreement with the H_{k2} value obtained for the studied Fe_3Si film. Since the direction of the uniaxial anisotropy φ_2 almost coincides with the direction of steps, it is quite probable that the main source of the uniaxial anisotropy in the Fe_3Si film on the vicinal substrate Si(111) is the magnetic dipole interaction.

Of particular interest is the unidirectional anisotropy observed in Fe_3Si films. Although the value $H_{k1} = 0.15$ Oe is almost an order of magnitude lower than the uniaxial anisotropy field, its contribution is noticeably manifested in the angular dependence of the resonance field. The unidirectional anisotropy was observed earlier in Co films grown epitaxially on Cu(1117) vicinal surfaces [24, 25]. The unidirectional shift measured using the magneto-optic Kerr effect of the hysteresis loop was about 1.5 Oe. Interestingly, this shift was observed at an angle of 65° to the step edges on the Co surface. In Fe_3Si films, we observed a similar picture: the unidirectional anisotropy is directed not along and not across the steps but at an angle of about 62° with respect to the direction orthogonal to the steps. The authors of [26] pointed to the possible physical nature of the unidirectional anisotropy in Co/Cu (1117). They believe that the most probable origin of the observed effect is the Dzyaloshinskii—Moriya interaction [27, 28] arising because of symmetry breaking on the steps of the ferromagnetic film. However, additional studies are necessary for the confirmation of this hypothesis.

It is important to note that the magnetic parameters of the phenomenological model of the studied film on the singular and vicinal surfaces have a relatively slight difference (see table). However, allowance in the model for the slight misorientation of the (111) substrate surface by the angle $\delta = 0.14°$ in the $[11\bar{2}]$ direction and by the angle $\xi = 5°$ in the $[\bar{1}10]$ direction leads to a considerable change in the angular dependence $H_R(\varphi_H)$ and to the almost exact correspondence between theory and experiment.

To summarize, the magnetic properties of the Fe_3Si epitaxial thin film grown on the Si(111) vicinal silicon surface have been studied using the FMR method. The effective saturation magnetization M_{eff} = 1105 G and the cubic magnetic anisotropy constant $K_4 = 1.15 \times 10^5$ erg/cm^3 have been determined. It has been established that the film also has uniaxial magnetic anisotropy characterized by the constant K_2 = 796 erg/cm^3 and slight unidirectional magnetic anisotropy with the constant K_1 = 163 erg/cm^3. The analysis of the results obtained has indicated that uniaxial magnetic anisotropy has a magnetic dipole nature, which is associated with the formation of magnetic charges on the roughnesses (steps) on the film surface existing because of the slight misorientation of the substrate with respect to the (111) surface. The high sensitivity of the FMR method to small deviations of the film surface of the (111) singular face makes it possible to use it for fast and relatively simple diagnostics of the extent of the misorientation of vicinal surfaces.

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