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Determination of magnetic anisotropies and miscut angles in epitaxial thin films on vicinal (111) substrate by the ferromagnetic resonance

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ABSTRACT

A method for determining magnetic anisotropy parameters of a thin single-crystal film on vicinal (111) substrate as well as substrate miscut angles from angular dependence of ferromagnetic resonance field has been proposed. The method is based on the following: (i) a new approach for the solution of the system of nonlinear equations for equilibrium and resonance conditions; (ii) a new expression of the objective function for the fitting problem. The study of the iron silicide films grown on vicinal Si(111) substrates with different miscut angles confirmed the efficiency of the method. The proposed method can be easily generalized to determine parameters of single-crystal films grown on substrates with an arbitrary cut.

1. Introduction

Epitaxial ferromagnetic films and various multilayer structures grown on single-crystal substrates are widely studied because of prospects of their application in spintronic devices [1,2]. Particular attention has recently been focused on the study of magnetic films grown on the vicinal cuts of Si(111) single-crystal substrates. For these substrates, methods of creating stepwise surfaces providing a high accuracy of the step width and step height are well developed [3,4]. This offers the possibility to control the magnetic properties of the films in a wide range by varying the miscut angle of the vicinal Si(111) surface in a narrow range [5,6]. In this context, methods for precise determining magnetic anisotropy parameters of the thin films [7] assume greater importance, among which ferromagnetic resonance (FMR) is a convenient and powerful technique [8].

Recently, we have pointed to the high sensitivity of the FMR technique to small miscut angles of a vicinal (111) surface in single-crystal thin films [9]. The possibility of azimuthal and polar miscut angles determination from FMR field angular dependences was demonstrated. The aim of this work is to describe in more detail our developed method for determining magnetic anisotropy parameters of a thin film and substrate miscut angles as well as the new approaches realized in it.

2. Theoretical background

Let us consider a thin film in an external in-plane magnetic field **H**. The equilibrium direction of the magnetization **M** and the ferromagnetic resonance condition for a thin-film sample can be obtained from the free energy density expression of the system

$$F(\theta, \phi) = -M_s H \cos(\phi - \phi_H)\sin\theta + F^a(\theta, \phi), \quad (1)$$

where the first term of Eq. (1) is the Zeeman energy contribution, and the second term is the magnetic anisotropy energy contribution. In the coordinate system *xyz* where the *z* axis coincides with the film normal, θ denotes the polar angle between the magnetization and the *z* axis, while φ and φ_H correspond to the azimuthal angles of **M** and **H** respectively, measured with respect to the *x* axis. M_s is the saturation magnetization. For a single-crystal thin ferromagnetic film deposited on a vicinal (111) surface with a small miscut angle δ (a model of the film is shown in Fig. 1), the density of the magnetic anisotropy energy can be written as [9]

$$\begin{aligned} F^a(\theta, \phi) = {} & 2\pi M_s^2 \cos^2\theta - K_\perp \cos^2\theta - \\ & - K_1 \sin\theta \cos(\phi - \phi_1) - K_2 \sin^2\theta \cos^2(\phi - \phi_2) + \\ & + K_4\left(\tfrac{1}{3}\cos^4\theta + \tfrac{1}{4}\sin^4\theta\right) - \tfrac{K_4\sqrt{2}}{3}\sin^3\theta\cos\theta\cdot\sin 3(\phi - \phi_4) + \\ & + \delta\tfrac{K_4}{6}\left\{\begin{array}{r} \sin 2\theta\cdot(7\sin^2\theta - 4)\cdot\sin(\phi - \phi_4 + \xi) + \\ +\sqrt{2}\sin^2\theta\cdot(7\sin^2\theta - 6)\cdot\cos 2(\phi - \phi_4 - \xi/2) - \\ -\sqrt{2}\sin^4\theta\cdot\cos 4(\phi - \phi_4 + \xi/4) \end{array}\right\} \end{aligned} \quad (2)$$

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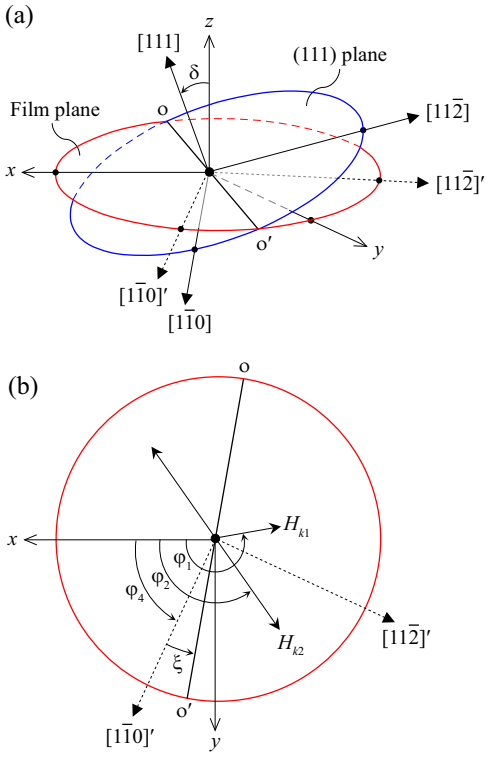

Fig. 1. The model and the orientation scheme of the crystallographic plane (111) of the single-crystal film with respect to its surface (a). Notations used in the phenomenological model of the thin film (top view) (b).

Here, the first term describes the energy contribution from the demagnetizing field of the film. The second term describes the energy of the uniaxial perpendicular anisotropy with the constant $K_\perp$. This magnetic anisotropy is associated with the symmetry breaking at the film surface and at the interface between the film and the substrate [7]. The following two terms of Eq. (2) describe the energies of the unidirectional (with constant K_1) and the uniaxial (with constant K_2) magnetic anisotropies, which fields H_{k1} and H_{k2} lie in plane of the film and are directed at angles φ_1 and φ_2 respectively (Fig. 1b). The remaining terms of the expression are related to the magnetocrystalline cubic anisotropy with the constant K_4 and the orientation φ_4 of the $[1\bar{1}0]'$ crystallographic direction (see Fig. 1).

In low-dimensional systems such as thin films, a shape anisotropy energy is usually the dominant term in the total magnetic anisotropy energy. The shape anisotropy is the main reason of in-plane orientation of the magnetization in the sample. The reorientation of the spontaneous magnetization from the film plane to the normal because of the surface anisotropy is possible only for ultrathin films with thicknesses of a few atomic layers [10]. Therefore, when the film is magnetized by the in-plane external magnetic field, the equilibrium angle θ equals π/2.

Using the Smith and Suhl formula [11,12], the ferromagnetic resonance equation and equilibrium condition can be written as follows

$$\left[H_R\cos(\phi_M-\phi_H)+\frac{F^a_{\phi\phi}}{M_s}\right]\times\left[H_R\cos(\phi_M-\phi_H)+\frac{F^a_{\theta\theta}}{M_s}\right]-\frac{F^{a\,2}_{\theta\phi}}{M_s^2}=\left(\frac{\omega_0}{\gamma}\right)^2, \tag{3}$$

$$H_R\sin(\phi_M-\phi_H)+F^a_\phi/M_s=0, \tag{4}$$

where $f_0=\omega_0/2\pi$ is the microwave pump frequency, and γ is the gyromagnetic ratio. The partial derivatives of $F^a_{\phi\phi}$, $F^a_{\theta\theta}$, $F^a_{\theta\phi}$, and F^a_ϕ have to be taken at the equilibrium position of the magnetization vector, that is, for angles $\theta=\pi/2$ and $\varphi=\varphi_M$, for which the total free energy density F has its minimum value.

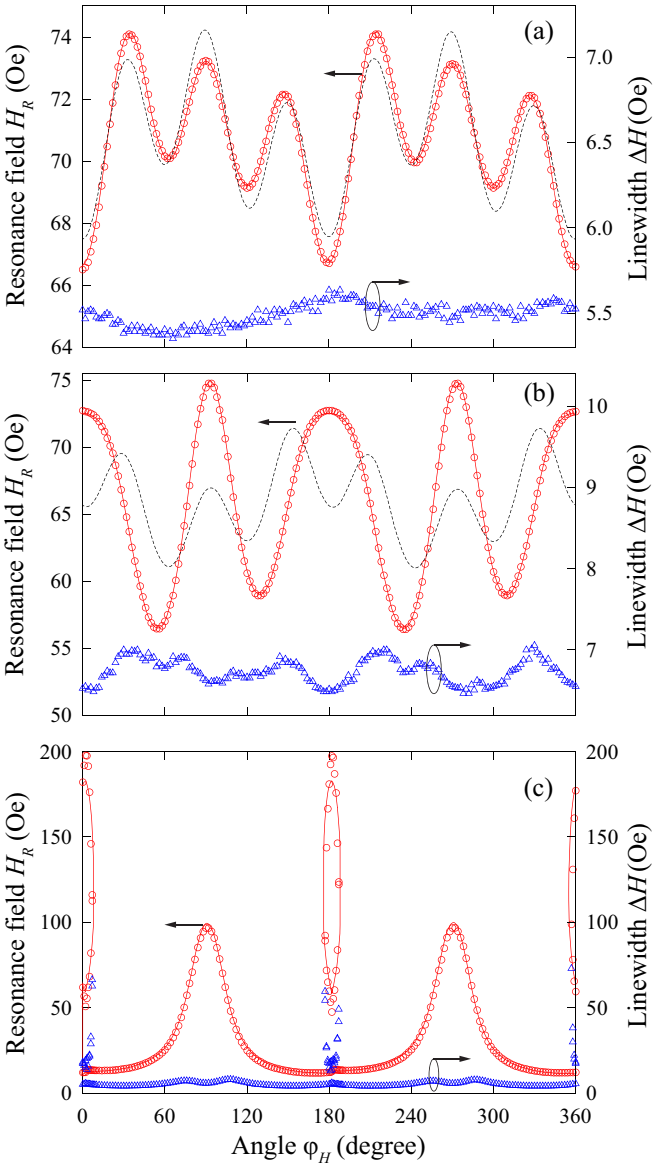

Fig. 2. Dependences of the resonance filed H_R and FMR linewidth ΔH on the sweeping magnetic field direction φ_H: (a) sample 1, (b) sample 2, (c) sample 3. Symbols correspond to the experimental measurements. Solid lines are the theoretical calculations for the model of a film on the vicinal surface with δ≠0, while dashed lines show the theoretical results for the case of singular surface (δ=0°).

In practice, methods for determination of model parameters by means of FMR are based on one or other numerical procedure that allows to approximate the experimental angular dependence $H^*_R(\phi_H)$ by the theoretical curve $H_R(\varphi_H)$, which is calculated from the nonlinear Eqs. (3) and (4). The practical realization of such a procedure faces two problems that influence both the accuracy and the reliability of the obtained results.

The first problem is directly related to the realization of an algorithm for simultaneous solution of the nonlinear equations for equilibrium and resonance conditions. When modeling the resonance field prior knowledge of the external field is required to calculate the equilibrium direction of **M**. The solution of this problem with a large number of unknown parameters for the considered here model leads not only to a long calculation time but also to an emergence of errors that are difficult to control. We suggest a quite simple solution of this problem, which can be used in all cases when a magnetization vector and an external magnetic field lie in the same plane. Let us first rewrite the system of the nonlinear Eqs. (3)–(4) as

Table 1
Optimal parameters of the thin film model for the three iron silicide samples. These parameters are obtained from the FMR field angular dependences by the suggested method.

	M_{eff} (G)	$H_{k1}=K_1/M_{eff}$ (Oe)	φ_1 (deg)	$H_{k2}=2K_2/M_{eff}$ (Oe)	φ_2 (deg)	$H_{k4}=2K_4/M_{eff}$ (Oe)	φ_4 ($\pm n\pi/3$, deg)	$\alpha=\varphi_4+\xi$ ($\pm n\pi/3$, deg)	δ (deg)
sample 1									
1	1 590.7	0.04	−39.6	1.21	156.3	297.35	30.7	48.4	0.097
2	1 583.1	0.04	40.0	1.11	157.0	296.95	31.0	48.2	0.098
	1 586.9	**0.04**	**0.2**	**1.16**	**156.6**	**297.15**	**30.9**	**48.3**	**0.097**
sample 2									
1	1 697.8	0.02	250.6	1.01	44.5	352.09	31.8	28.9	0.62
2	1 694.9	0.02	376.8	1.51	36.2	343.74	31.9	29.3	0.64
	1 696.3	**0.02**	**313.7**	**1.26**	**40.4**	**347.91**	**31.8**	**29.1**	**0.63**
sample 3									
1	1 490.2	0.28	54.0	31.86	93.7	498.76	30.7	27.6	3.93
2	1 491.5	0.14	229.0	28.70	95.6	498.53	29.7	23.1	3.77
	1 490.9	**0.21**	**141.5**	**30.28**	**94.6**	**498.64**	**30.2**	**25.4**	**3.85**

$$H_R\cos(\phi_M-\phi_H)=f(\phi_M)\equiv-\frac{F^a_{\phi\phi}+F^a_{\theta\theta}}{2M_s}+\frac{\sqrt{(F^a_{\phi\phi}-F^a_{\theta\theta})^2+4(F^a_{\theta\phi})^2+4M_s^2(\omega_0/\gamma)^2}}{2M_s},$$
$$H_R\sin(\phi_M-\phi_H)=g(\phi_M)\equiv-F^a_{\phi}/M_s. \tag{5}$$

If we transform these expressions to the complex representation and denote $Z(\varphi_M)=f(\varphi_M)+ig(\varphi_M)$ and $Z^*(\varphi_M)=f(\varphi_M)-ig(\varphi_M)$, we obtain

$$H_R=[Z(\phi_M)Z^*(\phi_M)]^{1/2},\quad e^{-i\phi_H}=e^{-i\phi_M}\left[\frac{Z(\phi_M)}{Z^*(\phi_M)}\right]^{1/2}. \tag{6}$$

Alternatively, in exponential form of the complex function $Z(\phi_M)=|Z(\phi_M)|e^{i\arg Z(\phi_M)}$

$$\phi_H=\phi_M-\arg Z(\phi_M),\qquad H_R=|Z(\phi_M)|. \tag{7}$$

Therefore, we obtain two independent equations. The first equation enable us to determine fast and accurate all equilibrium directions of the magnetization φ_M by using a simple iteration algorithm. The second equation allow us to calculate all values of the resonance fields.

The second problem is related to the choice of an objective function form f_{obj} for the realization of an algorithm that minimizes deviation between experimental and theoretical curves. The usage of the commonly accepted expression $f_{obj}=[H^*_R(\phi_H)-H_R(\varphi_H)]^2$ does not always enable to determine the optimal model parameters, and the final result can depend significantly on the choice of the initial values. For justification of the choice of our suggested objective function form let us write an approximate expression $f(\varphi_M)$ from (5). Taking into account the following notions

$$H_0=\frac{1}{4\pi M_{eff}}\left(\frac{\omega_0}{\gamma}\right)^2,\quad \sigma=\frac{H_0}{4\pi M_{eff}},\quad \lambda=-\frac{1}{4\pi M_{eff}}\frac{F^a_{\theta}}{M_s},$$
$$\varepsilon=\frac{1}{4\pi M_{eff}}\left[\frac{F^a_{\theta\theta}}{M_s}-4\pi M_{eff}\right],\quad \eta=\frac{1}{4\pi M_{eff}}\frac{F^a_{\theta\phi}}{M_s}, \tag{8}$$

where $M_{eff}=M_s-K_\perp/2\pi M_s$ is an effective saturation magnetization, $f(\varphi_M)$ in the linear approximation with respect to the small parameters ε, η, and λ will be

$$\begin{aligned}f(\phi_M)\approx{}& H_0(\omega_0)(1-\sigma(\omega_0))-\\ &-\frac{K_1}{M_s}\cos(\phi_M-\phi_1)-\frac{2K_2}{M_s}\left[1-\frac{1}{2}\frac{H_0}{4\pi M_{eff}}\right]\cos 2(\phi_M-\phi_2)+\\ &-\delta\frac{2\sqrt{2}}{3}\frac{K_4}{M_s}\left[1+\frac{K_4/M_s+3H_0}{4\pi M_{eff}}\right]\cos 2(\phi_M-\phi_4-\xi/2\pm\pi/2)-\\ &-\delta\frac{8\sqrt{2}}{3}\frac{K_4}{M_s}\left[1+\frac{K_4/M_s-3H_0/4}{4\pi M_{eff}}\right]\cos 4(\phi_M-\phi_4+\xi/4)-\\ &-2\frac{K_4^2/M_s^2}{4\pi M_{eff}}\cos 6(\phi_M-\phi_4\pm\pi/6)\end{aligned} \tag{9}$$

Several key points can be concluded from the analysis of the last expression. First, the dependence $f(\varphi_M)$—in contrast to the angular dependence of the resonance field $H_R(\varphi_H)=f(\varphi_M)/\cos(\varphi_M-\varphi_H)$—has a simple form and contains only several main terms of a Fourier expansion versus the equilibrium magnetization angle φ_M. From the above reasoning, we propose the following form of the objective function to solve fitting problem $f_{obj}=[H^*_R\cos(\phi_M-\phi_H)-f(\phi_M)]^2$. This choice, as will be demonstrated in Section 3, enable us to determine effectively parameters of the theoretical model, including cases when angular dependences $H^*_R(\phi_H)$ with several resonance fields values for certain angles are obtained. Second, the Eq. (9) is convenient to use for evaluation of the initial values of the model parameters. Third, the Eq. (9) enables six equivalent directions of the angle $\phi_4=\phi'_4\pm n\pi/3$ (where n is integer) and, therefore, six equivalent azimuthal miscut angles $\alpha=\varphi_4+\xi$. This means that we cannot determine α unambiguously from the analysis of the resonance field dependence. However, the direction of the azimuthal miscut angle can be chosen based on general physical considerations. For example, in Ref. [9] it was demonstrated that this could be done by an analysis of the direction φ_2 of the uniaxial magnetic anisotropy formed in the film.

3. Experiment and discussion

For testing of our proposed method, we have studied three samples of epitaxial iron silicide thin films. The samples were prepared by simultaneous thermal evaporation of iron and silicon from two crucibles in ultra-high vacuum (1.3×10^{-8} Pa) and subsequent deposition of atoms on the boron-doped atomically clean vicinal Si(111) substrate (see Ref. [13] for details). The films were grown on vicinal Si(111) surfaces with the following nominal miscut angles: 0.1° for the first sample, 0.6° for the second sample, and 4° for the third sample. Magnetic properties of the produced samples were studied by the automated scanning spectrometer of ferromagnetic resonance [14,15] at the pump frequency f_0 =3.329 GHz. For each sample, FMR angular dependences were measured on two local areas (~0.8 mm^2) with a spacing of 4 mm.

Fig. 2 shows angular dependences of the resonance field H_R(circles) and FMR linewidth ΔH (triangles) measured on one local area of each sample. In this figure, theoretical angular dependences are also shown (solid lines), with model parameters obtained from the experimental dependences $H^*_R(\phi_H)$ by our developed method. Here, a good agreement between theory and experiment is clearly seen. To illustrate the significance of the substrate miscut influence on the resonance field angular dependences, in Fig. 2a and b we additionally plotted theoretical dependences (dashed lines) calculated for a model of a film on the singular (111) surface with δ=0° and, as in the previous cases, for optimal parameters of the model. These results demonstrate that even such small misorientation as $\delta\approx0.1°$ and $\delta\approx0.6°$ of the samples 1 and 2 surfaces from the singular (111) surface leads to the significant

modification of the resonance curves behavior.

The calculated parameters of the measured samples are presented in Table 1. The parameters of different local areas (1 and 2 in Table 1) within each sample are in good accordance, whereas some discrepancies can be seen between averaged parameters of the samples (shown as bold font in Table 1). This might be related to a slight change in the stoichiometric composition of the investigated films. However, the obtained values of the polar miscut angle δ of the vicinal Si(111) surface are in reasonably good agreement with the nominal values.

4. Conclusion

In conclusion, we would like to emphasize that the proposed method can be easily generalized to determine parameters of single-crystal films grown on substrates with an arbitrary cut. Particularly, in our recent work [16] we have successfully adapted this method for the accurate determination of parameters of an in-plane magnetic anisotropy with an arbitrary number of expansion terms.

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References

[1] S.A. Wolf, D.D. Awschalom, R.A. Buhrman, J.M. Daughton, S. von Molnar, M.L. Roukes, A.Y. Chtchelkanova, D.M. Treger, Science 294 (2001) 1488 http://dx.doi.org/10.1126/science.1065389.
[2] I. Zutic, J. Fabian, S.D. Sarma, Rev. Mod. Phys. 76 (2004) 323 http://dx.doi.org/10.1103/RevModPhys.76.323.
[3] J.-L. Lin, D.Y. Petrovykh, J. Viernow, F.K. Men, D.J. Seo, F.J. Himpsel, J. Appl. Phys. 84 (1998) 255 http://dx.doi.org/10.1063/1.368077.
[4] F.K. Men, F. Liu, P.J. Wang, C.H. Chen, D.L. Cheng, J.L. Lin, F.J. Himpsel, Phys. Rev. Lett. 88 (2002) 096105 http://dx.doi.org/10.1103/PhysRevLett.88.096105.
[5] A. Stupakiewicz, E.Y. Vedmedenko, A. Fleurence, T. Maroutian, P. Beauvillain, A. Maziewski, R. Wiesendanger, Phys. Rev. Lett. 103 (2009) 137202 http://dx.doi.org/10.1103/PhysRevLett.103.137202.
[6] K.S. Ermakov, Yu.P. Ivanov, L.A. Chebotkevich, Phys. Solid State 52 (2010) 2555 http://dx.doi.org/10.1134/S1063783414100114.
[7] Y. Shiratsuchi, M. Yamamoto, S.D. Bader, Prog. Surf. Sci. 82 (2007) 121 http://dx.doi.org/10.1016/j.progsurf.2006.08.001.
[8] M. Farle, Rep. Prog. Phys. 61 (1998) 755 http://dx.doi.org/10.1088/0034-4885/61/7/001.
[9] B.A. Belyaev, A.V. Izotov, JETP Lett. 103 (2016) 41 http://dx.doi.org/10.1134/S0021364016010033.
[10] M.T. Johnson, P.J.H. Bloemen, F.J.A. Den Broeder, J.J. De Vries, Rep. Prog. Phys. 59 (1996) 1409 http://dx.doi.org/10.1088/0034-4885/59/11/002.
[11] J. Smit, H. Beljers, Philips Res. Rep. 10 (1955) 113.
[12] H. Suhl, Phys. Rev. 97 (1955) 555 http://dx.doi.org/10.1103/PhysRev.97.555.2.
[13] I.A. Yakovlev, S.N. Varnakov, B.A. Belyaev, S.M. Zharkov, M.S. Molokeev, I.A. Tarasov, S.G. Ovchinnikov, JETP Lett. 99 (2014) 527 http://dx.doi.org/10.1134/S0021364014090124.
[14] B.A. Belyaev, A.V. Izotov, A.A. Leksikov, IEEE Sens. 5 (2005) 260 http://dx.doi.org/10.1109/JSEN.2004.842293.
[15] B.A. Belyaev, A.A. Leksikov, I.Ya Makievskii, V.V. Tyurnev, Instrum. Exp. Tech. 40 (1997) 390.
[16] B.A. Belyaev, A.V. Izotov, P.N. Solovev, Physica B 481 (2016) 86 http://dx.doi.org/10.1016/j.physb.2015.10.036.