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Structural and Magnetic Characteristics of Fe/Si Bilayer and Multilayer Films Obtained by Thermal Deposition in Ultrahigh Vacuum

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Abstract—The structural and magnetic characteristics of Fe/Si bilayer and multilayer films with nanometer-thick layers obtained by thermal deposition in ultrahigh vacuum have been studied by methods of small-angle X-ray scattering, electron spectroscopy, and magnetometry. It is established that the mechanisms involved in the formation of Fe/Si and Si/Fe interfaces are different. © *2005 Pleiades Publishing, Inc.*

In recent years, increasing attention has been devoted to multilayer magnetic structures in view of their unique physical properties and good prospects for practical applications [1, 2]. In particular, the phenomenon of colossal magnetoresistance, which had originally been discovered in metal superlattices, was later also observed in multilayer structures composed of alternating metal and semiconductor layers [3]. Investigations into the properties of multilayer structures with semiconductor interlayers (spacers) open the way to the creation of novel materials possessing unique properties for micro- and nanoelectronics [4]. In the technology of multilayer structures consisting of ultrathin layers, there are special requirements related to precise control over the composition of each layer, which implies that the deposition of layers has to be performed under ultrahigh vacuum (UHV) conditions.

This Letter presents the results of investigation of the structural and magnetic characteristics of Fe/Si bilayer and multilayer films with nanometer-thick layers obtained by thermal deposition in UHV on silicon substrates.

We have studied multilayer structures comprising iron layers with silicon spacers obtained by thermal evaporation in vacuum with deposition on single crystal Si(100) and Si(111) substrates at room temperature. The samples were prepared on a modified molecular beam epitaxy setup of the Angara type with a residual vacuum in the UHV technological chamber 10^{-7} Pa. The component materials were evaporated from refractory (boron nitride) crucibles. The evaporation rate could be controlled by varying the crucible temperature, so that the deposition rates were equal to 0.16 nm/min for iron and 0.9 nm/min for silicon. The control over the temperatures of Fe and Si evaporators and their shutters used for the formation of multilayer structures was provided by an automated programmed complex [5].

The compositions of deposited layers were studied by Auger electron spectroscopy (AES) and electron energy loss spectroscopy (EELS). These measurements

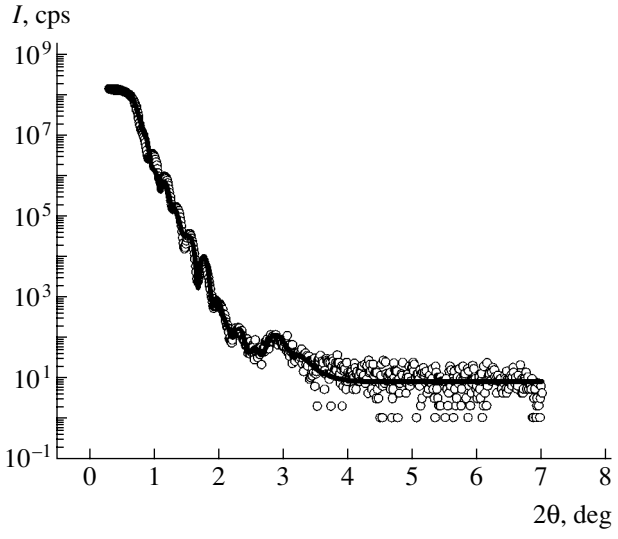

Fig. 1. The typical experimental SAXS data (open symbols) and the results of model calculations (solid curves) for a 20-layer [Fe(2 nm)/Si(2 nm)] × 9/Fe(10 nm)/Si(10 nm) structure.

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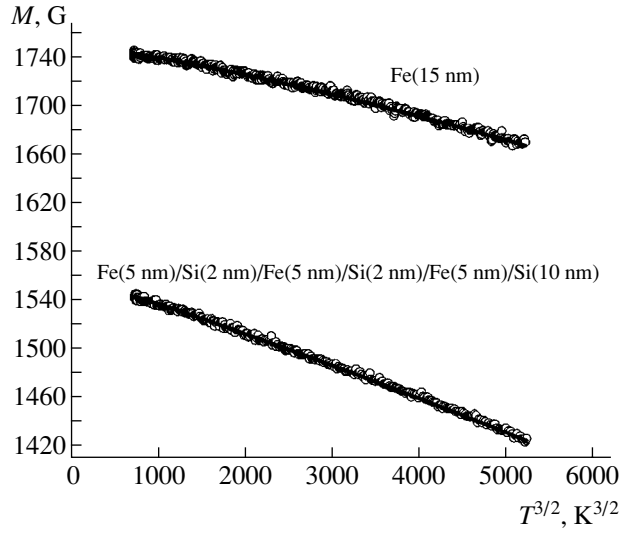

Fig. 2. The typical curves of the magnetization M versus temperature T (on the $T^{3/2}$ scale) for a [Fe(5 nm)/Si(2 nm)] × 2/Fe(5 nm)/Si(10 nm) superlattice measured in an external magnetic field of H = 1 kOe in the temperature range from 4.2 to 250 K: open circles present the experimental data and solid curves show the results of their approximation using expression (1).

were performed *in situ* in the UHV technological system. The structural characteristics (layer thickness, interface roughness) of [Fe(2 nm)/Si(2 nm)] × 9/Fe(10 nm)/Si(10 nm) superlattices were determined from the results of small-angle X-ray scattering (SAXS) measurements [6]. Figure 1 presents the typical experimental SAXS data (open symbols) and the results of model calculations (solid curves) for this structure. Table 1 gives the characteristics of Fe and Si layers obtained from the model calculations, which provided for the best fit to the experimental SAXS data for a 20-layer [Fe(2 nm)/Si(2 nm)] × 9/Fe(10 nm)/Si(10 nm) structure, in comparison to the values determined using technological parameters. The values of the surface roughness determined by fitting the model SAXS intensity profiles to the measured data were comparable with the thicknesses of the component layers. The absolute values of roughness were independent of the thicknesses of adjacent Fe and Si layers and of the order (Fe/Si or Si/Fe) of their deposition. These data characterize the inhomogeneity of multilayer structures with respect to the thicknesses of Fe and Si layers.

Table 1. Data on the layer thickness determined from the deposition time ($d_{technol}$) and the thickness (d_{calcd}) and roughness (h_{calcd}) calculated from the SAXS data for a 20-layer [Fe(2 nm)/Si(2 nm)] × 9/Fe(10 nm)/Si(10 nm) structure

Layer	$d_{technol}$, nm	d_{calcd}, nm	h_{calcd}, nm
Si	10	9.87	2.16
Fe	10	13.71	1.94
Si	2	1.46* ± 0.27	1.34* ± 0.16
Fe	2	1.51* ± 0.27	1.46* ± 0.21
Si	Substrate	–	1.62

* Average for 9 bilayers.

The magnetic properties of Fe/Si multilayers were studied on an automated vibrating-sample magnetometer with a superconducting coil [7]. Figure 2 shows the typical curve of the magnetization M versus temperature T (on the $T_3/2$ scale) for a [Fe(5 nm)/Si(2 nm)] × 2/Fe(5 nm)/Si(10 nm) superlattice measured in an external magnetic field of H = 1 kOe in the temperature range from 4.2 to 250 K. The general character of this $M(T)$ curve is indicative of the absence of paramagnetic and superparamagnetic responses. As can be seen from Fig. 2, the experimental data are well described in terms of a theoretical relation known as the Bloch law (depicted by solid lines) [8]:

$$M(T) = M_0(1 - BT^{3/2} - CT^{5/2}), \quad (1)$$

where M_0 is the zero-temperature magnetization and B and C are temperature-independent coefficients. A relation of the B value to the main magnetic characteristics of the material is described by the empirical formula

$$A = \frac{k}{8\pi}\left(\frac{M_0}{g\mu_B}\right)^{1/3}\left(\frac{2.612}{B}\right)^{2/3}, \quad (2)$$

where A is the exchange coupling constant and μ_B is the Bohr magneton.

Table 2 presents the A, B, and C constants for the multilayer Fe/Si structures under consideration in comparison to the data for body-centered cubic (bcc) α-Fe crystals and a single iron film on silicon. The coincidence of the values of magnetization for a single Fe layer and a bulk bcc α-Fe crystal shows that this Fe layer was also composed of pure bcc iron. A decrease in the effective exchange constant A for this single-layer iron film (with a thickness of d = 15 nm) as determined from the Bloch $T^{3/2}$ law in comparison to the value for bcc α-Fe is consistent with the well-known effect of decreasing effective exchange predicted for thin films within the framework of the spin wave theory [9].

The decrease in M_0 and A values observed for a [Fe(5 nm)/Si(2 nm)] × 2/Fe(5 nm)/Si(10 nm) multilayer film is related to the mutual penetration of Fe and Si atoms into the neighboring layers of this structure [10]. This interdiffusion results in the formation of an interfacial region composed of a Fe–Si solid solution or iron silicides. The observed increase in B (or a decrease in A) and a decrease in M_0 for iron layers of smaller thickness can be readily explained. These changes are related to the formation of a magnetic het-

erophase system and, hence, correspond to an increase in the volume fraction of the interfacial region with lower values of the exchange constant and magnetization. The magnetization of the iron layer with such an interfacial region can be described as

$$M_0 = M_b n_b + M_a n_a = M_b - (M_b - M_a)\frac{2\Delta}{d_{\mathrm{Fe}}}, \quad (3)$$

where n_a and n_b are the fractions of Fe atoms in the interfacial regions and iron layers, respectively, and M_a and M_b and the corresponding magnetizations; Δ is the thickness of the interfacial region; and d_{Fe} is the iron layer thickness. Relation (3) corresponds to a simplified model, whereby a concentration profile characteristic of the real Fe–Si interface is replaced by a stepwise profile [11] including a transition layer with a constant composition and the thickness Δ. In terms of such a model Fe–Si interface with a constant composition and the effective magnetic parameters M_a and A_a, it is convenient to evaluate Δ characterizing the thickness or volume fraction of the interfacial region.

An analysis of the phase diagram of the Fe–Si system [12, 13] showed that the interfacial regions of a multilayer Fe/Si structure can feature the formation of a ferromagnetic solid solution or Fe_3Si and FeSi compounds. For obtaining estimates, we suggested that $M_a = M(Fe_3Si = 1270$ G [12] (nonmagnetic silicide was rejected, since its presence could not account for the observed decrease in the exchange coupling constant A). Within the framework of this model, we have evaluated the thickness of weakly-magnetic interfacial regions formed in a six-layer [Fe(5 nm)/Si(2 nm)] × 2/Fe(5 nm)/Si(10 nm) structure. The total thickness of interfaces in this structure was estimated at 6Δ = 5.4 nm.

Assuming that the thicknesses of transition layers at the Fe/Si and Si/Fe interfaces are the same, the thickness of each interfacial region in the six-layer [Fe(5 nm)/Si(2 nm)] × 2/Fe(5 nm)/Si(10 nm) structure under consideration is Δ = 0.9 nm. In this case, a single Si/Fe interface for the aforementioned 15-nm-thick Fe film on a silicon substrate must, according to formula (3), decrease the magnetization by 30 G. However, the data in Table 2 show that no such decrease takes place for a real iron film on silicon substrate. This discrepancy can be related to a difference in the parameters of Fe/Si and Si/Fe junctions, which, in turn, is caused by the different mechanisms of formation of such interfaces in the course of sample preparation by means of thermal deposition.

In order to elucidate the mechanism of the Fe–Si interface formation, we have studied by EELS the bilayer structures of two types, Fe(10 nm)/Si(1.5 nm) and Si(10 nm)/Fe(1.5 nm), which were specially prepared for this purpose. It was found that the electron energy loss spectrum of a Si(10 nm)/Fe(1.5 nm) sample with the upper layer of iron is virtually the same as that of a single 10-nm-thick iron film on a silicon substrate. In contrast, the spectrum of a sample with the upper layer of silicon exhibited peaks with energy positions (21.9 eV for the first bulk plasmon) corresponding to those reported for iron Fe_3Si silicide [14]. Our results also agree with the conclusions made by Klasges *et al.* [15] upon a comparison of the electronic structures of iron silicides formed in a Fe film on Si substrate and a Si film on Fe substrate. According to the data of spin-resolved photoemission spectroscopy, it was concluded that the mechanisms of formation of Si/Fe and Fe/Si interfaces are different [15]: in one case, a metal-like interface with a thickness of ~2 nm and a composition close to Fe_3Si was formed, whereas the photoemission spectrum in the other case corresponded to an amorphous layer of bulk silicon or a Fe_xSi_{1-x} so lid solution with small x.

Table 2. The main magnetic characteristics of multilayer Fe/Si structures

Material	M_0, G	B, 10^{-5} K$^{-3/2}$	C, 10^{-8} K$^{-5/2}$	A, 10^{-6} erg/cm
α-Fe (fcc)	1740	0.34	0.1	2.1
Single Fe layer	1750	0.49	0.1	1.7
Multilayer Fe/Si structure	1560	1.38	1.0	0.9

In conclusion, the results of our complex study of the structural and magnetic characteristics of Fe/Si multilayer films obtained by thermal deposition in UHV on single crystal silicon substrates show evidence for a significant influence of the transition region with a modified chemical composition formed at the Fe–Si interface on the magnetic properties of the system. The possible difference in the mechanisms of formation of the Fe/Si and Si/Fe interfaces must be taken into consideration in the interpretation of physical properties of multilayer Fe–Si systems.

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REFERENCES

1. M. N. Baibich, J. M. Broto, A. Fert, *et al.*, Phys. Rev. Lett. **61**, 2472 (1988).
2. R. E. Cambley and R. L. Stamps, J. Phys.: Condens. Matter. **5**, 3727 (1993).
3. A. E. Berkowitz, J. R. Mitchell, M. J. Carey, *et al.*, Phys. Rev. Lett. **68**, 3745 (1992).
4. V. P. Dragunov, I. G. Neizvestnyĭ, and V. A. Gridchin, *Grounds of Nanoelectronics: A Textbook* (Izd. NGTU, Novosibirsk, 2004) [in Russian].

5. S. N. Varnakov, A. A. Lepeshev, S. G. Ovchinnikov, *et al.*, Prib. Tekh. Éksp. **6**, 252 (2004).
6. E. E. Fullerton, I. K. Schuller, H. Vanderstraeten, and Y. Bruynserade, Phys. Rev. B **45**, 9292 (1992).
7. A. D. Balaev, *Physics of Magnetic Films: Collection of Scientific Works* (Irkutsk, 1980), Vol. 14, p. 171 [in Russian].
8. F. Keffer, in *Handbuch der Physik*, Vol. XVIII.2: *Ferromagnetism*, Ed. by H. P. J. Wijn (Springer, Berlin, 1966).
9. D. L. Mills and A. A. Maradudin, J. Phys. Chem. Solids **28**, 1855 (1967).
10. G. J. Strijkers, J. T. Kohlhepp, H. J. M. Swagten, and W. J. M. de Jonge, Phys. Rev. B **60**, 9583 (1999).
11. R. S. Iskhakov, N. A. Shepeta, S. V. Komogortsev, *et al.*, Fiz. Met. Metalloved. **95** (3), 37 (2003).
12. *Magnetic Properties of Metals: D-Elements, Alloys and Compounds*, Ed. by H. P. J. Wijn (Springer, Berlin, 1991), p. 190.
13. C. J. Smithells, *Metals Reference Book* (Butterworths, London, 1967), Vol. 2.
14. J. M. Gallego and R. Miranda, J. Appl. Phys. **69**, 1377 (1991).
15. R. Klasges, C. Carbone, W. Eberhardt, *et al.*, Phys. Rev. B **56**, 10801 (1997).

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