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MAGNETISM AND FERROELECTRICITY

Size Effects and Magnetization of $(Fe/Si)_n$ Multilayer Film Nanostructures

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Abstract—The temperature dependence of the magnetization of $(Fe/Si)_n$ multilayer films with nanometer layers is investigated. The films are prepared through thermal evaporation under ultrahigh vacuum onto Si(100) and Si(111) single-crystal substrates. It is revealed that the thickness of individual iron layers in $(Fe/Si)_n$ multilayer films affects the magnetization and its temperature dependence. The inference is made that this dependence is associated with the formation of a chemical interface at the Fe–Si boundaries. The characteristics of the chemical interface in the $(Fe/Si)_n$ films are estimated.

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1. INTRODUCTION

Considerable research interest in magnetic multilayer nanostructures is associated with their unique physical properties and prospects for practical applications [1–3]. Magnetic multilayers and sandwich structures with alternating semiconductor and metal nanolayers have attracted the particular attention of researchers, because their properties can be modified over a wide range through controllable introduction of different impurities, exposure to radiation of different types, and variation in temperature [4–9]. Iron–silicon film nanostructures exhibit different phenomena, including a temperature-dependent change in the exchange interaction parameter [7, 10] and a photoinduced change in the interlayer exchange interaction [4, 11, 12].

In the majority of theoretical models concerning the aforementioned physical properties, it is assumed that interfaces between layers in nanostructures are continuous and sharp [7]. However, it is known that, in similar materials with nanometer thicknesses of individual layers, the roughness of interfaces between neighboring layers and interdiffusion of their components are important types of structural imperfection and can substantially affect the physical properties [13]. As a consequence, the physical parameters of multilayer nanostructures should depend on the thickness of individual layers. In this respect, the separation of contributions from the roughness of neighboring layers and chemical interface to the physical properties of materials is an important problem. Since the saturation magnetization (or the magnetic moment per atom) and the exchange constant are additive quantities that are sensitive to the nearest environment of atoms, their investigation makes it possible to separate correctly the contributions from the chemical interface and the layer roughness [14].

The purpose of this work is to investigate the influence of the thickness of individual iron layers on the magnetization and its temperature dependence, which allows one to estimate the exchange constant for $(Fe/Si)_n$ multilayer film nanostructures.

2. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUE

Samples for our investigations were prepared by thermal evaporation onto Si(100) single-crystal substrates under ultrahigh vacuum at room temperature on a modified Angara molecular-beam epitaxy setup [15]. The base pressure in a technological vacuum chamber was equal to 10^{-7} Pa. The materials were evaporated from high-temperature boron nitride crucibles. The growth rate of layers of the corresponding materials was controlled in situ with the use of LÉF-751M fast laser ellipsometer and amounted to 0.3 nm/min for Fe and 1.4 nm/min Si. Subsequently, the layer thicknesses were accurately determined ex situ by x-ray fluorescence spectroscopy. The temperature of the evaporators and operation of the shutters during the preparation of the multilayer structures were controlled with an automated software–hardware system [15]. The chemical composition of the layers prepared was monitored in situ using Auger electron and characteristic electron energy loss spectroscopy [15]. Analysis of the small-angle x-ray scattering curves of the $(Fe/Si)_n$ multilayer films demonstrated that the materials prepared have a well-defined superlattice whose characteristic modulation periods are in reasonable agreement with the specified technological parameters [14].

The magnetic characteristics of the $(Fe/Si)_n$ multilayer films were measured on a SQUID magnetometer in fields up to 50 kOe at temperatures in the range from 4.2 to 400 K. In the absence of a field, the magnetic moment lies in the film plane. This manifested itself in characteristic rectangular hysteresis loops obtained in the geometry in which the external field was parallel to the film plane. The coercive forces of the samples under investigation varied from 60 to 200 Oe. The temperature dependences of the magnetization were measured in an external field $H = 700$ Oe for the samples at saturation. The magnetic properties measured on the SQUID magnetometer were compared with those determined in our earlier work [14] with the use of a vibrating-coil magnetometer.

3. RESULTS

The temperature dependences of the normalized magnetization measured for the multilayer structures in an external field $H = 0.7$ kOe are plotted in Fig. 1. The general behavior of the dependences $M(T)$ obtained in the temperature range from 4.2 to 400 K indicates the absence of a superparamagnetic response.

It can be seen from Fig. 1 that the slope of the temperature dependence of the magnetization increases with a decrease in the Fe layer thickness. The general relationship that describes the decrease in the magnetization of a ferromagnetic metal with an increase in the temperature can be written in the form [16]

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

or as the expansion in the dimensionless temperature

$$M(T) = M_0(1 - b(T/T_C)^{3/2} - d(T/T_F)^2 - c(T/T_C)^{5/2} - \dots), \quad (1a)$$

where M_0 is the saturation magnetization at 0 K. The terms $BT^{3/2}$ and $CT^{5/2}$ take into account the contribution from the excitation of thermal magnons with the dispersion law $\omega \sim 1 - \cos(\pi k/a)$ to the decrease in the magnetization [16]. The term DT^2 in relationship (1) is associated with the Fermi excitations of electrons in metals [17]. The approximation of the experimental dependences $M_e(T)$ by relationship (1) with the use of the least-squares procedure leads to the appearance of negative fitting parameters D or C. This has no physical

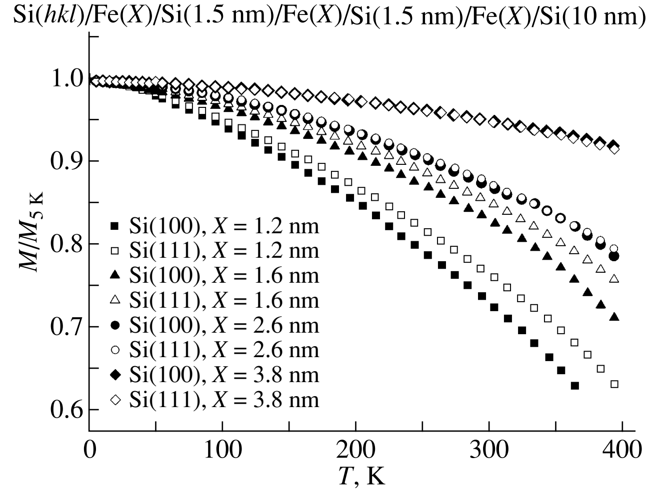

Fig. 1. Temperature dependences of the normalized saturation magnetization $M/M_{5\,K}$ for $(Fe/Si)_n$ multilayer films at different thicknesses of Fe(X) individual layers.

meaning and is associated with the large number of terms in the approximating function (1). The representation of the dependence $M(T)$ in the form of expression (1a) permits us to evaluate the contributions of different terms to the temperature dependence of the magnetization. The dimensionless coefficients b, c, and d for ferromagnets are of the order of unity [16, 17]. The Curie temperature for iron can be estimated as $T_C \sim 10^3$ K. The Fermi temperature for metals is $T_F \geq 10^4$ K. As a result, by choosing $T = 200$ K (the average temperature in our measurements), we obtain $(T/T_C)^{3/2} \sim 10^{-1}$, $(T/T_C)^{5/2} \sim 10^{-2}$, and $(T/T_F)^2 \sim 10^{-4}$. These estimates enable us to eliminate the term associated with the Fermi excitations. Hereafter, the experimental curves $M(T)$ will be described by the relationship that accounts for the decrease in the magnetization with the increase in the temperature only due to the excitation of thermal spin waves; that is,

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

The dependences of the quantities M_0, B, and C on the thickness of individual iron layers were calculated using the approximation of the experimental dependences $M(T)$ (Fig. 1) by expression (2). These dependences and the data obtained in our previous work are depicted in Figs. 2–4. It can be seen from Figs. 2 and 3 that a decrease in the thickness of the individual iron layer leads to a considerable decrease in the magnetization M_0 and an increase in the Bloch constant B. The dependence of the constant C on the thickness t_{Fe} exhibits a nonmonotonic behavior: as the thickness of the individual iron layer decreases, the constant C increases to $t_{Fe} = 2$–3 nm and then sharply decreases in the range $t_{Fe} = 1.1$ nm.

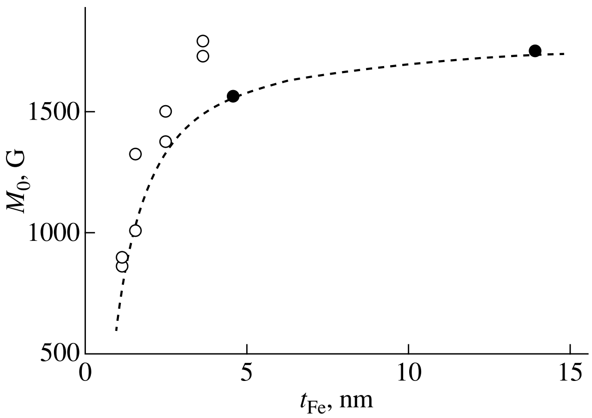

Fig. 2. Dependence of the magnetization M_0 for $(Fe/Si)_n$ multilayer films on the thickness of the Fe individual layer. Open circles indicate the data obtained in this work, and closed circles correspond to the data taken from [14].

4. DISCUSSION

The origin of the decrease in the magnetization M_0 with the decrease in the Fe layer thickness in the $(Fe/Si)_n$ films can be associated with the change in the contribution made to the magnetization by the intermediate layer (interface) that is formed at the boundary between the Fe and Si layers and has a lower magnetization [14, 18, 19]. The formation of this layer can be caused, for example, by the penetration of Fe and Si atoms into the neighboring layers of the multilayer film [14, 18, 19]. This results in the formation of the interlayer phase, which is a solid solution of Fe–Si compounds.

The change in the magnetization M_0 due to the magnetic heterophase structure of the system is described by the relationship

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

where n_a and n_b are the fractions of surface Fe atoms (entering into the interface) and bulk Fe atoms, respectively; M_b is the magnetization of the Fe layer; M_a is the magnetization of the surface phase; and Δ is the thickness of the interface, which is the Fe–Si alloy (Fig. 5). This linear dependence represents the model simplification of the observed situation where the real concentration profile, which characterizes the alloy of a varying composition in the interface region, is replaced by a stepwise concentration profile (whose parameters, i.e., the composition and the interface thickness Δ, characterize the alloy of constant composition [14, 18]). The model of the Fe–Si alloy, which has a constant composition in the interface region and possesses the magnetization M_a, makes it possible to estimate the volume fraction of this alloy or the interface thickness Δ. The dependence of the experimental magnetization on the quantity t_{Fe}^{-1} (Fig. 6) is in good agreement with relationship (3). This suggests the validity of the aforementioned heterophase model and allows us to use relationship (3) for evaluating the interface parameters. The interface parameters estimated from the data presented in Fig. 6 are as follows: $0 < M_a < 850$ G and $0.7 < 2\Delta < 1.1$ nm. According to relationship (3), the lower limit of the interface thickness corresponds to $M_a = 0$. There-

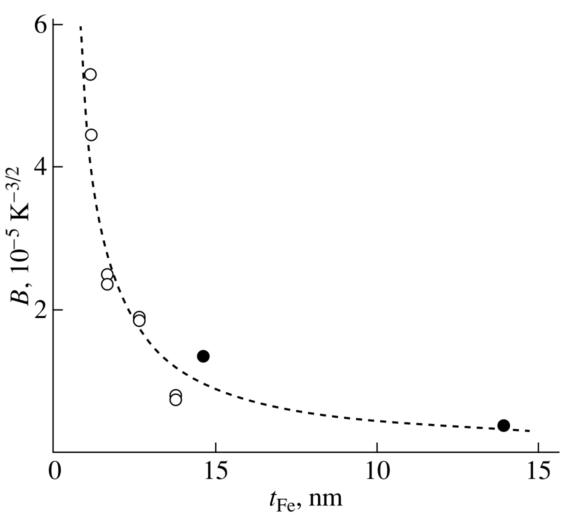

Fig. 3. Dependence of the Bloch constant B for $(Fe/Si)_n$ multilayer films on the thickness of the Fe individual layer. Open circles indicate the data obtained in this work, and closed circles correspond to the data taken from [14].

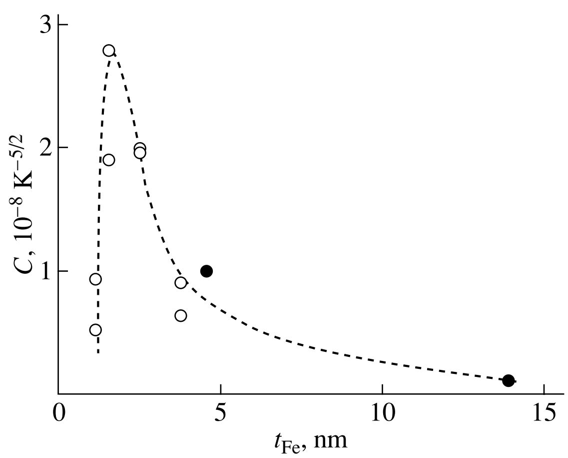

Fig. 4. Dependence of the constant C for $(Fe/Si)_n$ multilayer films on the thickness of the Fe individual layer. Open circles indicate the data obtained in this work, and closed circles correspond to the data taken from [14].

fore, the question as to whether the Fe–Si alloy layer possesses a spontaneous magnetization cannot be answered uniquely from the data presented in Fig. 6. The determined thickness of the chemical interface ($\Delta \sim 0.4$ nm) is considerably smaller than the thickness of the morphological interface (roughness) estimated for our films by small-angle x-ray diffraction (~1.8 nm) [14]. The data taken from [20] on the magnetization of $(Fe/Si)_n$ films prepared by ion-beam sputtering are also presented in Fig. 6. In this case, the estimation of the interface parameters leads to the following results: $0 < M_a < 1000$ G and $1.6 < 2\Delta < 2.5$ nm. Consequently, the interface thickness in the $(Fe/Si)_n$ films studied in [20] is larger than that in our films by a factor of 1.5–2.0. Analysis of the data available in the literature on the structure and composition of alloy interfaces at boundaries between Fe and Si layers demonstrates that stable phases (Fe–Si magnetic solid solutions [12, 21–24], as well as nonmagnetic ε-FeSi and β-Fe_2Si silicides [12, 23, 24]) and metastable silicides (Fe_3Si [12, 21], α-$FeSi_2$ [12, 23], *c*-FeSi [12, 22, 23]) can be formed at the Fe/Si interface. The magnetization of the heterophase interface is an averaged quantity, because it is determined by the phase composition and magnetization of the components. Strijkers et al. [22] showed that, for Si layers up to 1.5 nm thick, the intermediate layer between the Fe layers in Fe–Si multilayers is formed in the form of crystalline monosilicide FeSi (metastable phase with a structure of the CsCl type) with a lattice epitaxially matching to the neighboring Fe layers. The possibility of forming a Fe–Si semiconductor layer with a spontaneous magnetization is of considerable interest for spintronics problems, because this layer should lead to spin polarization of electrons when the electric current flows through a magnetic heterostructure.

5. EXCHANGE INTERACTION CONSTANT

The coefficient B in relationship (1) and the main magnetic constants of a material, namely, the exchange interaction constant A and the magnetization M_0, are related by the formula [16]

$$A = \frac{k}{8\pi}\left(\frac{M_0}{g\mu_B}\right)^{1/3}\left(\frac{2.612}{B}\right)^{2/3}. \quad (4)$$

The calculated exchange interaction constants A for the $(Fe/Si)_n$ films under investigation are presented in Fig. 7. The coincidence of the exchange interaction constant for the Fe single-layer film 14 nm thick with the corresponding constant for bulk chemically pure iron with a body-centered cubic structure indicates that the this Fe layer consists of chemically pure iron with a body-centered cubic structure. This is in complete agreement with the data on the magnetization, which, for the Fe single-layer film 14 nm thick, is equal to the magnetization of bulk chemically pure iron with the body-centered cubic structure. A decrease in the effective exchange interaction constant for the $(Fe/Si)_n$ mul-

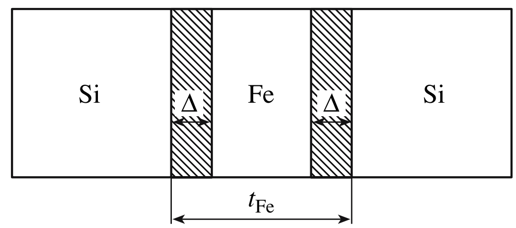

Fig. 5. Magnetic heterophase model of the $(Fe/Si)_n$ system.

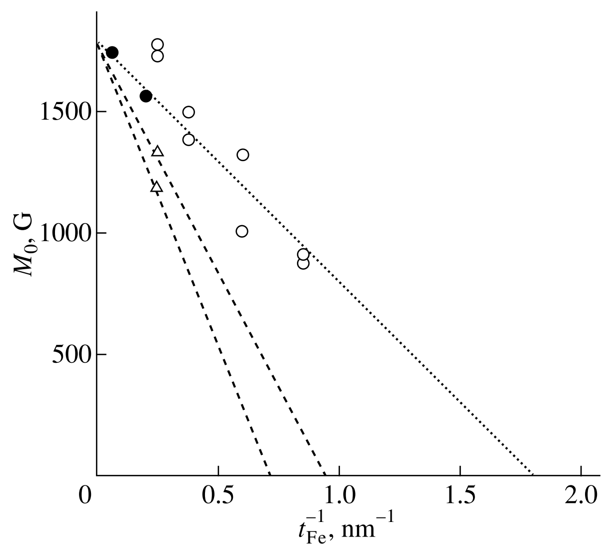

Fig. 6. Dependence of the experimental magnetization for $(Fe/Si)_n$ multilayer films on the quantity t_{Fe}^{-1}. Open circles indicate the data obtained in this work, closed circles correspond to the data taken from [14], and triangles represent the data taken from [20].

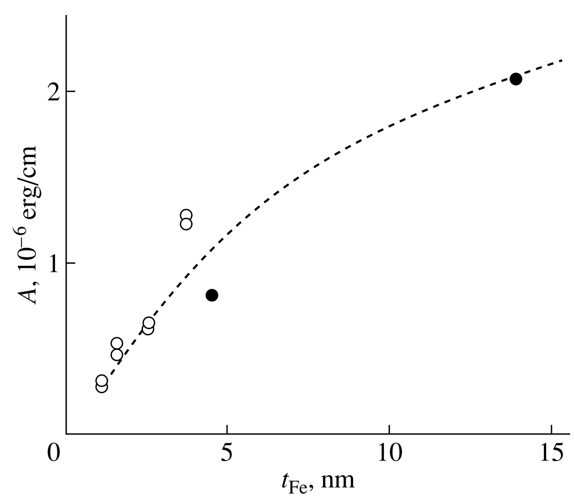

Fig. 7. Dependence of the exchange interaction constant A for $(Fe/Si)_n$ multilayer films on the thickness t_{Fe}^{-1} of magnetic layers according to calculations from formula (4). Open circles indicate the data obtained in this work, and closed circles correspond to the data taken from [14].

tilayer films under investigation is inconsistent with the weakening of the effective exchange predicted for thin films within the spin-wave theory [25]. Actually, according to Mills and Maradudin [25], the ratio between the Bloch constant B_s for an ultrathin ferromagnetic layer and the Bloch constant B_b for a bulk material is $B_s/B_b = 2$. Correspondingly, the ratio between the exchange interaction constants [see formula (4)] should be $A_s/A_b = 1/2^{2/3} = 0.63$. This means that the effective exchange interaction constant in relationships (1) and (4) for ultrathin layers can decrease by no more than 37% as compared to that for the bulk material. It can be seen from Fig. 7 that the exchange interaction constant A changes by a factor of more than seven with a variation in the Fe layer thickness in the films under investigation. For multilayer structures, the effective exchange interaction constant determined from the temperature dependence of the magnetization according to formulas (1) and (4) should depend on the partial effective exchange interaction constants A_i (characterizing the Fe and interface layers) and on the exchange interaction that occurs between the ferromagnetic layers through the Si layers. Moreover, the exchange interaction constant turns out to be very sensitive to different defects in the atomic structure of the ferromagnetic layer [18, 26]. In this respect, the discussion of theoretical expressions appropriate for interpreting the data presented in Fig. 7 is beyond the scope of the present paper.

6. CONCLUSIONS

Thus, we investigated the low-temperature dependence of the magnetization for $(Fe/Si)_n$ multilayer films with nanometer layers. It was revealed that the magnetization and the exchange interaction constant estimated from the temperature dependence of the magnetization for the $(Fe/Si)_n$ films decrease considerably with a decrease in the thickness of the individual iron layer from 10 to 1 nm. The conclusion was drawn that this behavior is associated with the magnetic interface layer that is formed at the Fe–Si boundaries and possesses magnetic parameters differing from those of the Fe layer. The use of the heterophase model of the structure of the $(Fe/Si)_n$ layered nanosystem made it possible to estimate some characteristics, such as the thickness, magnetization, and exchange interaction constant, for the interface region in the $(Fe/Si)_n$ films.

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