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LOW-TEMPERATURE MAGNETISM

Spontaneous magnetization and characteristics of temperature-induced magnetization of planar Co/Si nanostructures

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The magnetic properties of planar Co/Si nanostructures with different nominal thicknesses of the magnetic (2–42 nm) and nonmagnetic (0.3–10 nm) constituent layers are studied in the temperature range 4.2–300 K. It is established that in the presence of Si layers the spontaneous magnetization of Co decreases and its temperature dependence changes, and magnetic hysteresis is modified. The interlayer influence is interpreted as being due to the diffusion of Si into the Co layers, which results in the formation of magnetically disordered boundary interfaces with low average magnetization. The depth of the interfaces depends on the nominal thickness of the Si layers and is estimated to reach 1.6 nm. A qualitative explanation of the characteristics of the magnetization of the experimental objects under the influence of a magnetic field and temperature is given in a model where the interfaces have a granular microstructure. Electron-microscope observations confirm some assumptions of the proposed model. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2720079]

INTRODUCTION

Cobalt belongs to the class of magnets that are actively used for developing heterogeneous functional media. These include planar superlattices[1] and granular Co/Si films with gigantic magnetoresistance,[2] Co/SiO_2 mulitlayers[3] with a gigantic Hall effect, Co/Pt (Ref. 4) and Tb/Co (Ref. 5) multilayer films for recording information, and others. Various physical properties of Co are used in these materials. However, they all rely, one way or another, on the specific nature of its electronic system, which on the one hand is characterized by high spin polarization and strong ferromagnetism and on the other hand manifests a strong tendency to undergo a transformation in the presence of other chemical elements. The latter property is especially important for nanosize layered structures where Co can lose its individuality under the influence of the adjoining layers having a different composition and exhibit degradation of functional properties. An effect of this kind, expressed as a decrease of the average atomic magnetic moment, has been recorded in, specifically, ultrathin Co/Si films.[6,7] The present work is devoted to a study of such objects on the basis of measurements and analysis of the temperature dependences of the magnetic properties of planar Co/Si nanostructures with layers of different thicknesses.

EXPERIMENTAL SAMPLES AND PROCEDURE

Multilayer Co/Si films obtained by high-frequency ionic sputtering in an Ar atmosphere at pressure 10^{-4} mm Hg and in the presence of a uniform magnetic field with intensity 100 Oe were investigated. The samples were formed on glass substrates by alternately sputtering different elements. The thicknesses of the Co (L_{Co}) and Si (L_{Si}) layers were determined according to the sputtering time with known rates of deposition of the indicated materials. These rates were found as a result of a preliminary experiment and were 0.07 nm/s for Co and 0.03 nm/s for Si with error ~10%. The presence of a magnetic field in the gas-discharge zone produced a uniaxial magnetic anisotropy in the plane of the films. A sampling analysis of the microstructure was performed with an electron microscope on samples deposited on NaCl and freed from the substrates immediately before the investigation. A SQUID magnetometer was used for measuring the magnetic properties.

PRESENTATION AND ANALYSIS OF THE EXPERIMENTAL RESULTS

Figure 1 shows (dots) the values of the reduced average magnetic moment of Co atoms ($<\mu_{Co}>$) in films with different thicknesses of the Co layers and a fixed thickness of the Si layers (L_{Si}=2 nm). Such a structure can be briefly described by the expression $[Co(L_{Co})/Si(2)]_n$, where n is the number of periods. The values of L_{Co} and n were varied in the course of the experiment from 2.1 to 42 nm and from 20 to 2 nm, respectively, so that the total thickness of the magnetic component was 80–40 nm. The values of $<\mu_{Co}>$ were calculated from measurements of the saturation magnetization of the films at T=4.2 K. The normalization quantity, was the saturation magnetization of a sample with L_{Co}=42 nm.

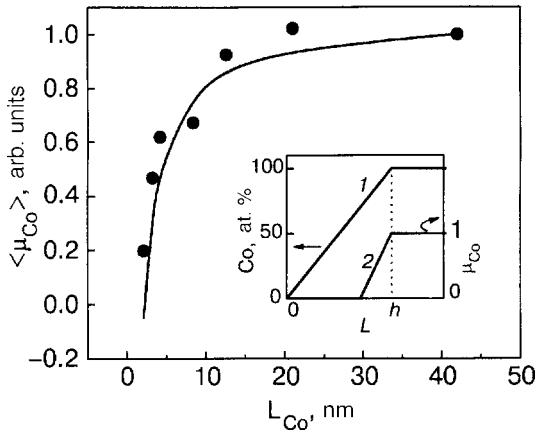

FIG. 1. Reduced values of the average magnetic moment of Co atoms in $[Co(LCo)/Si(2)]_n$ films versus the thickness of the Co layers: dots—experimental data; line—computed curve for a model structure with interface thickness $h=1.2$ nm. The inset illustrates the model of a Co–Si interlayer interface of thickness h with linear variation of the composition along the thickness (1) and the corresponding variation of the reduced magnetic moment of Co (2).

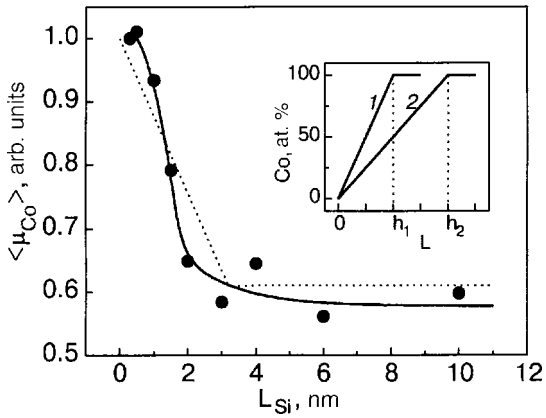

FIG. 2. Experimental (dots, solid line) and computed (dotted line) dependences of the reduced value of the average magnetic moment of the Co atoms in $[Co(7)/Si(L_{Si})]_9/Co(7)$ films versus the thickness of the Si layers. The inset illustrates schematically the possible change of the configuration of the interlayer interface with a change in the nominal thickness of the Si layer: small (1), large (2).

The data presented show that a strong decrease of the atomic magnetic moment of Co does indeed occur in samples with a sufficiently thin layered structure ($L_{Co}<10$ nm).This characteristic can be explained on the basis of the well-known fact that Si actively affects the magnetism of Co in amorphous Co–Si alloys,[8] and it is sometimes called the "charge-transfer effect." According to the data in Ref. 8 Si strongly, and linearly (as a function of the concentration), decreases the atomic magnetic moment of Co so that the alloy becomes virtually nonmagnetic when up 30 at. % impurity is introduced. It is entirely likely that interlayer boundary diffusion occurs in our objects and results in the formation of transitional regions (interfaces) with variable binary composition and low magnetization. The decrease in L_{Co} corresponds to an increase of the relative interfacial fraction in the total volume of the films, which, in turn, is expressed as a progressive decrease of $<\mu_{Co}>$.

The simple model shown schematically in the inset in Fig. 1 can be proposed to make a quantitative estimate of the thickness of the interfaces. Let the concentration of Co increase linearly from the surface into the interior volume of the magnetic layer and reach 100% at distance h (line 1). In addition, let the magnetic moment of Co be zero for Si concentration above 30 at.%, i.e. at depths up to $0.7h$, and increase linearly at large distances from the surface, reaching its maximum value at depth h (line 2). Then the relation between $<\mu_{Co}>$ and the thickness of Co can be determined by a simple analytic expression containing the parameter h:

$$<\mu_{Co}> = 1 - 2\left(\frac{0.7h}{L_{Co}} + \frac{0.3h}{2L_{Co}}\right) = 1 - 1.7\frac{h}{L_{Co}}. \qquad (1)$$

An example of the dependence $<\mu_{Co}>(L_{Co})$ calculated for $h=1.2$ nm and tied to the experimental data at the point $L_{Co}=42$ nm is presented in Fig. 1 (line). The indicated value of h was chosen using a fitting procedure. It gives the best computational curve $<\mu_{Co}>(L_{Co})$, so that in the present model it can be used as a qualitative characteristic of real interfaces.

To study the interlayer effect in Co/Si nanostructures we performed another experiment on the films $[Co(7)/Si(L_{Si})]_9/Co(7)$. According to this structural formula the thickness of the Co layers in this series of samples was constant ($L_{Co}=7$ nm), and the film structure contained 9 periods and one other Co surface layer. The thickness of the Si layers varied from one sample to another in the range 0.3–10 nm. Figure 2 shows (dots and solid line) the experimental dependence of the reduced average magnetic moment of Co on the thickness of the Si layers. The values of $<\mu_{Co}>$ were determined by the method described above but the normalizing quantity in this case was the saturation magnetization of a uniform Co film.

Two very different sections can be distinguished on the curve $<\mu_{Co}>(L_{Si})$ shown in Fig. 2: in the first section ($0<L_{Si}<3$ nm) $<\mu_{Co}>$ decreases sharply; on the second section ($L_{Si}>3$ nm) the magnetic moment is essentially constant. This behavior of $<\mu_{Co}>$ can be interpreted as follows. When the thickness of the nonmagnetic layers has the nominal value all of the Si goes to the formation of the interlayer interfaces, i.e. it dissolves in the Co layers. Obviously, $<\mu_{Co}>$ decreases as a result. Near $L_{Si}=3$ nm the interlayer diffusion saturates, and as L_{Si} increases further a real Si layer forms. An increase in the thickness of this layer, of course, has no effect on $<\mu_{Co}>$. To make quantitative estimates it can be assumed that at the formation stage the interfaces have a similar profile of the chemical composition: the Co content varies linearly from 0 up to 100%. At the same time different Si penetration depths correspond to different nominal values of L_{Si} (see inset in Fig. 2). In such a model, using the relation adopted above between the Si concentration and the atomic magnetic moment of Co, which is reflected in (1), the function $<\mu_{Co}>(L_{Si})$ can be calculated. It will have a break at a point corresponding to the maximum possible thickness h of the interfaces. The dotted line in Fig. 2 shows an example of such a dependence for the optimal value $h=1.6$ nm, found by a fitting procedure.

It is evident in Fig. 2 that the computed curve correctly conveys the characteristic features of the corresponding experimental dependence. Together with the results obtained

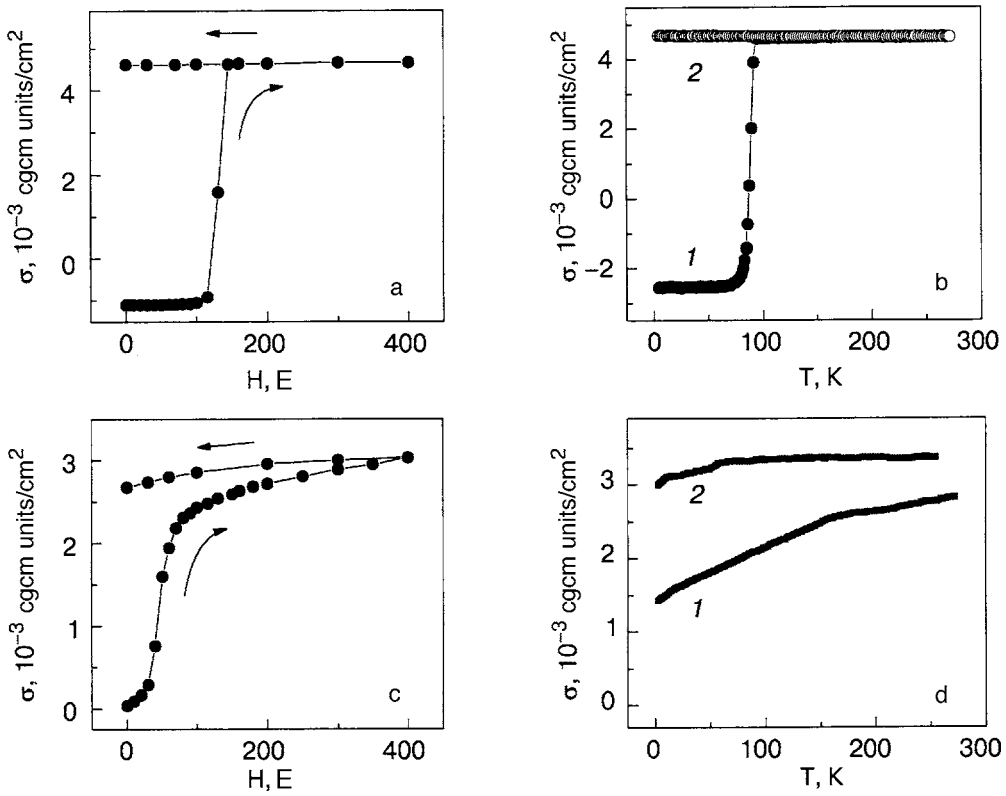

FIG. 3. Fragments of hystersis loops (a, c) and temperature dependences of the magnetization (b, d) in magnetic fields with intensities 50 Oe (1) and 400 Oe (2) for the samples $[Co(7)/Si(0.3)]_9/Co(7)$ (a, b) and $[Co(7)/Si(2)]_9/Co(7)$ (c, d).

for the samples $[Co(L_{Co})/Si(2)]_n$ this makes it possible to talk about the adequacy of the model where the interlayer interaction in the Co/Si system is interpreted in terms of interlayer interfaces. At the same time the results of the temperature study of Co/Si films showed that a simple continuum model does not reflect all features of the properties of these nanostructures. This refers, first and foremost, to the magnetic hysteresis, whose specific nature in $[Co(7)/Si(L_{Si})]_9/Co(7)$ films is demonstrated in Fig. 3.

Figures 3a and 3c show fragments of the hysteresis loops $\sigma(H)$ of films with different thickness of the Si interlayers. The quantity σ is the magnetic moment of a unit area of the samples. The measurements were performed at $T=4.2$ K in a magnetic field oriented along an induced axis of easy magnetization according to the scheme $0 \rightarrow H_{max} \rightarrow 0$. The samples were first demagnetized with a sign-changing field with decreasing amplitude. As one can see, the film with $L_{Si}=3$ nm (Fig. 3a) has a sharp threshold for magnetization reversal, which is due to, specifically, the fact that the method adopted does not completely demagnetize the film. A similar picture of magnetization reversal, which apparently occurs by abrupt motion of domain boundaries, is characteristic for all samples with $L_{Si} \leq 1$ nm. However, for thick nonmagnetic layers the situation changes qualitatively: magnetization occurs more evenly, the residual magnetization decreases appreciably (by 20–30%), and hysteresis appears in fields much stronger than the coercive force (Fig. 3c).

In our opinion the observed transformation is probably due to nonuniform interlayer diffusion. Suppose that Si penetrates into the fine-crystalline layers of Co predominantly along grain boundaries. Then the Co crystallites near the interfaces can be magnetically disordered along the depth. Their near-surface parts will be nonmagnetic because of the high enrichment with silicon and the interior parts will remain magnetically ordered. On the whole such a micro structure looks like a collection of magnetic particles (granules) in a Co–Si nonmagnetic matrix. Assuming the granules to be single-domain, the process of their magnetization can be characterized as a rotation of magnetization. This is what determines the decrease of the residual magnetization and the presence of high-field "tails" on the hysteresis loops of films with $L_{Si} \geq 1.5$ nm. However, apparently, the magnetization orientation in the central parts of the Co layers changes because of displacements of the domain walls, which is the main reason why the magnetization changes in weak fields.

The subtle nuances of the microstructure on which the model presented above was constructed are difficult to record directly. Nonetheless, some information about the real state of the microstructure can be gleaned from observations in an electron microscope. The results of such an investigation are presented in Fig. 4 in form of images of the microstructure and fragments of electron diffraction patterns of films of the type $[Co(L_{Co})/Si(2)]_5$. As one can see, the sample with $L_{Co}=10$ nm (Fig. 4a) is a fine-crystalline object whose structural components exhibit large size dispersion. Analysis of the diffraction pattern shows that these components are crystallites of hexagonal Co. Traces of other crystalline phases, for example Si or silicides of Co, were not seen in the films. The average size of the crystallites does not exceed 10 nm. Such crystallites, if they are magnetically isolated, indeed should be in a single-domain state, since even at room temperature the critical diameter of single-domainness for Co is greater

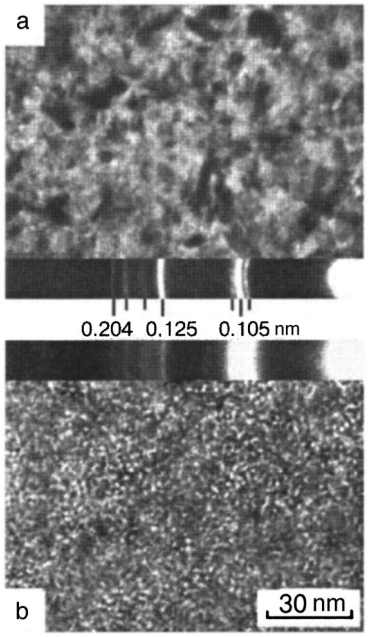

FIG. 4. Electron-microscope images of the microstructure and the corresponding electron diffraction patterns of the samples $[Co(10)/Si(2)]_5$ (a) and $[Co(3)/Si(2)]_5$ (b).

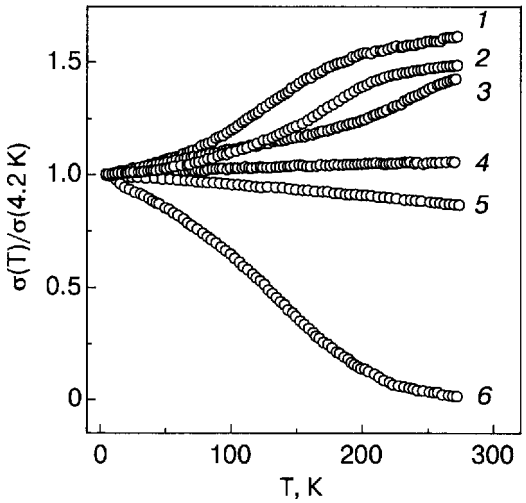

FIG. 5. Temperature dependences of the reduced specific magnetization of the films $[Co(L_{Co})/Si(2)]_n$ with Co layer thicknesses, nm: 42 (1), 12.6 (2), 8.4 (3), 4.2 (4), 3.2 (5), and 2.1 (6). The measurements were performed with the samples heated in a magnetic field $H_0=100$ Oe.

than 60 nm. In addition, the diffuseness of the diffraction lines shows that nanosize (1–2 nm) Co crystallites are present in the films; these crystallites can easily form the foundation of the interlayer interfaces.

Figures 3b and 3d show the temperature dependences of the specific magnetization for the films whose hysteresis loops were discussed above. These data were obtained by heating the samples in a constant magnetic field H_0 with different intensity. First they were cooled to $T=4.2$ K and demagnetized. Then, a field H_0 was applied along the easy magnetization axis, and the temperature was raised monotonically. The curves $\sigma(T)$ obtained in this manner also exhibit a relation with L_{Si}. When the nominal thickness of Si is small ($L_{Si}<1$ nm), the field $H_0=400$ Oe makes it possible to reach magnetic saturation in the entire temperature range employed (Fig. 3b, curve 1). A relatively weak field ($H_0=50$ Oe) has no effect on the magnetization up to some temperature, but above this temperature it causes an abrupt transition into the state of magnetic saturation (Fig. 3b, curve 2). Such *temperature-induced* magnetization is a consequence of a decrease of the coercive force, which, in turn, reflects the temperature behavior of the magnetic anisotropy of Co. The picture of "*thermal magnetization*" starts to change in samples with $L_{Si}=1.5$ nm and is found to be substantially different for $L_{Si}\geq 2$ nm (Fig. 3d). In this case the procedure used to obtain the initial state gives virtually complete demagnetization of the samples. However, in a weak field substantial magnetization is established even at $T=4.2$ K, and the curve $\sigma(T)$ is monotonically increasing right up to room temperature (curve 2). A gradual increase of the magnetization with increasing temperature is also observed in a strong field, but, it is true, in a much smaller temperature interval (curve 1).

The features noted above can also be given an interpretation using the idea of magnetic nonuniformity of the Co layers and their magnetization at the center, primarily as a result of the motion of domain walls, and at interfaces because of the rotation of magnetization. In addition, the formation of interfaces, in our opinion, results in a strong decrease of the coercivity of the domain walls. This is a well-known effect, which is observed in various layered objects where the exchange coupling between the magnetic layers breaks down.[9] It is due to the transition from through Néel domain walls to walls localized in individual layers but coupled by a magnetostatic interaction in a system with low energy density. Thus the magnetization which a sample with $L_{Si}=2$ nm exhibits at $T=4.2$ K and $H_0=50$ Oe (Fig. 3d, curve 2) can be ascribed to the interior part of the Co layers which are not touched by diffusion processes.

The temperature variation of σ is most likely a result of the interfaces and is due to the rotation of the magnetization in granules. As T increases it is facilitated because of a corresponding decrease of the magnetic anisotropy. The gradual nature of the increase of σ is due to the dispersion in the magnetic reorientation fields and can be due to several factors. In the first place, the particles have different sizes, which is due to the natural spread in the sizes of the Co crystallites and also the fact that Si diffuses to different depths in them. When it approaches the critical diameter of superparamagnetism the particle size starts to influence the coercive force directly. For Co this diameter, in order of magnitude, is 1–10 nm at temperatures below room temperature. In the second place, there is a dispersion of the spontaneous magnetization. This dispersion arises because of differences in the Si concentration in the Co granules. In the third place, possible oscillations of the magnetic anisotropy occur, also as a result of the concentration fluctuations. In addition, the magnetic anisotropy has a direct and mediated effect on the coercive force of the particles. In the latter case it is realized through the mechanism of superparamagnetism.

The factors determining the mechanisms of the thermal magnetization of Co/Si films include not only the thickness of Si but also the thickness of the magnetic layered component. The latter is demonstrated in Fig. 5, which shows the

temperature dependences of the reduced specific magnetization $\sigma(T)/\sigma(4.2\ K)$ for samples with different values of L_{Co}. These data were obtained using the above-described method in a magnetic field $H_0=100$ Oe. As one can see, all curves are monotonic but their character gradually changes as the L_{Co} decreases: they change from rising to falling. In samples of this series the thickness of the Si layers is large enough for interlayer interfaces, whose magnetization according to the model presented above gives rise to a smooth increase of the magnetization with increasing temperature, to form. Consequently, the course of the curves $\sigma(T)/\sigma(4.2\ K)$ in samples with relatively thick Co layers appears to be entirely natural.

The fact that the amplitude of the changes in the magnetization decreases and even becomes negative as L_{Co} decreases can be attributed to the rearrangement of the microstructure. This is indicated directly by studies in an electron-microscope. Returning to Fig. 4, which reflects the state of the samples with $L_{Co}=10$ nm (a) and $L_{Co}=3$ nm (b), it can be stated that a decrease of the thickness of the Co layers radically increases the dispersity of the structure. The average size of the regions of coherent scattering decreases to 2–3 nm. Nevertheless, the indications of the hexagonal crystalline structure of Co remain (see the corresponding electron diffraction pattern). On the basis of this result it can be supposed that even in interfaces the granule size decreases as a result of the strongly magnetic center. In granules, the volume fraction of the Co–Si solid solution with a zero or low magnetic moment on a Co atom increases, which naturally leads to a smaller change of the magnetization as a function of temperature. In addition, a decrease of the atomic magnetic moment reflects on the Curie temperature. This gives rise to a tendency for σ to decrease with temperature, and this trend starts to dominate in films with $L_{Co}\leq 3$ nm (Fig. 5, curves 5 and 6).

Our estimates show that in films with $L_{Co}=2.1$ nm all Co is located in very small granules with a strongly decreased Curie temperature. It is this circumstance that gives rise to the quite sharp temperature decrease of the magnetization and actual vanishing of σ near room temperature. It is interesting that in fields $H_0\leq 50$ Oe this sample exhibits a nonmonotonic variation of the magnetization with temperature. This is evident in Fig. 6, which shows the temperature dependences $\sigma(T)$ measured for different intensities of the field H_0. The course of the curves 1 and 2 can be explained by the fact that the previously described thermal magnetization of the granules prevails to the left of the maximum values of σ. In addition, it is quite intense, which shows that the magnetic anisotropy is low in these Co–Si formations. The temperature variation of the spontaneous magnetization of this material, which because of the low Curie temperature results in a sharp decrease of σ, becomes determining to the right of the maxima.

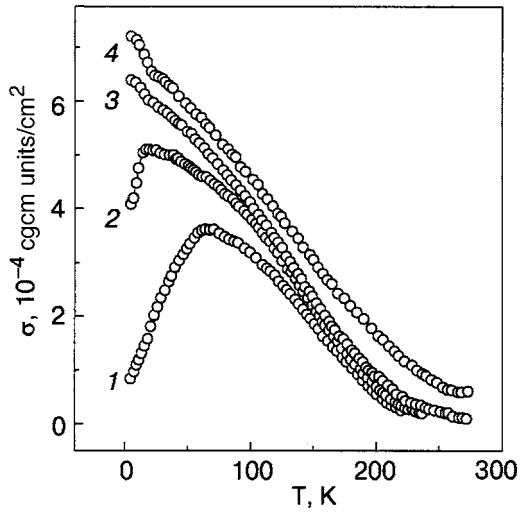

FIG. 6. Temperature dependences of the specific magnetization of the sample $[Co(2.1)/Si(2)]_{20}$, measured in magnetic fields with different intensities, Oe: 25 (1), 50 (2), 100(3), and 500 (4).

CONCLUSIONS

It was shown that the silicon in layered Co/Si structures strongly influences the magnetism of the Co layers. The effect depends on the thicknesses of the magnetic and nonmagnetic components and is expressed as a decrease of the average atomic magnetic moment and a specific change of the hysteresis characteristics. The model of interlayer interfaces with a granular magnetic structure gives a qualitative explanation of this behavior.

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