Nanostructure of Interfaces and Giant Magnetoresistance of Co/Cu Superlattices

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ABSTRACT

Specific features of the structure of interfaces in $[Co/Cu]_n$ superlattices obtained by magnetron sputtering have been studied by nuclear magnetic resonance (NMR). Modification of interface structural characteristics and magnetoresistive properties of the superlattices with the increase of the number of [Co/Cu] bilayers is analyzed. Correlation between magnetoresistance and interface structural characteristics has been revealed.

Keywords: giant magnetoresistance, superlattices, interfaces, nuclear magnetic resonance

1 INTRODUCTION

Modern magnetoresistive nanomaterials consisting of ultra-thin layers of magnetic and non-magnetic metals demonstrate an effect of giant magnetoresistance due to which they find wide application in many technical areas and attract great attention of the researchers [1]. One of the main problems in this rapidly developing field of research is characterization of the structural perfection of multilayered films, including their interface topology. In most studies the low-angle X-ray reflectometry is applied to characterize the state of interfaces [2, 3]. However, when fitting the X-ray spectra, a great number of parameters (up to 30) are varied, and the results of the spectra treatment are not always unambiguous.

Good opportunities to probe the state of layers and interfaces are provided by the Mossbauer spectroscopy [4-7], but application of this method is limited, since as a rule a Fe layer (preferably, enriched with the ^{57}Fe isotope) is required. Much wider spectrum of multilayers can be studied by the nuclear magnetic resonance (NMR) [8]. Particularly, NMR on ^{59}Co nuclei was used to study magnetic properties and structural features of $[Co/Cu]_n$ superlattices [9, 10].

The main idea in the application of this technique for the studies of $[Co/Cu]_n$ superlattices is that due to the hyperfine interaction magnetic moments of cobalt electrons create local magnetic fields at the ^{59}Co nuclei location, the magnitude and direction of which strongly depend on magnetic and structural features of the nearest surrounding of a probe-nucleus. Since the NMR enables to investigate the local field distributions in a specimen, it is a powerful tool for the research of structural characteristics and magnetic properties of various multilayered film systems. Particularly, the analysis of NMR spectra on ^{59}Co nuclei can provide information on the interface state and quality in $[Co/Cu]_n$ superlattices.

The Co/Cu superlattices demonstrate the record-breaking values of magnetoresistance among the magnetic metal materials [11, 12]. The goal of the present research is to reveal the correlation between the number of bilayers, magnetoresistive properties and the state of interfaces in Co/Cu superlattices.

2 EXPERIMENT

The $[Co(1.5nm/Cu(0.9nm)]_n$ superlattices (where n = 10, 20, 30 or 40) were manufactured by DC magnetron sputtering in the Ulvac MPS-4000-C6 device on glass substrates. To protect the superlattice surfaces from oxidation the specimens were covered with a Cr layer 3 nm thick. The buffer layer of Fe was 5 nm thick.

The superlattices were sputtered at room temperature and fixed argon pressure of 0.1 Pa, the magnetron power being 100 W. With these conditions the Fe, Co, Cu and Cr targets could be sputtered at the rates of 2.7, 3.0, 6.9 and 3.0 nm/min, respectively. Before sputtering the substrate surfaces were purified by ion etching in argon atmosphere in the sputtering device. The sputtering rate and surface quality were determined and controlled by the noncontact method of scanning interferometry.

Magnetoresistance was measured by a standard DC four-contact technique with the current flowing in the layers plane. Magnetic field was applied perpendicular to the current in a film plane. All the measurements were carried out at room temperature. The magnetoresistance was determined as follows: MR = $\Delta R/R_s \cdot 100\%$, $R_s = (R(H)-R_s)/R_s \cdot 100\%$, where R_s is the resistance in the magnetic saturation field.

Figure 1 demonstrates field dependences of magnetoresistance of superlattices with 10 and 40 bilayers, possessing the magnetoresistance of 38 and 24 % respectively.

The structure of superlattices was studied by transmission electron microscopy (TEM) in Philips CM30 electron microscope.

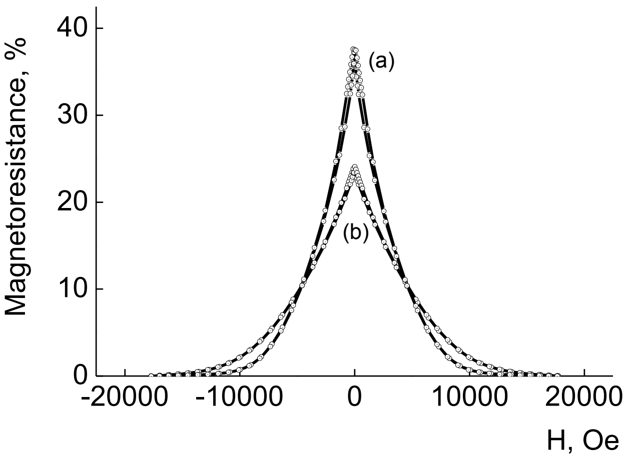
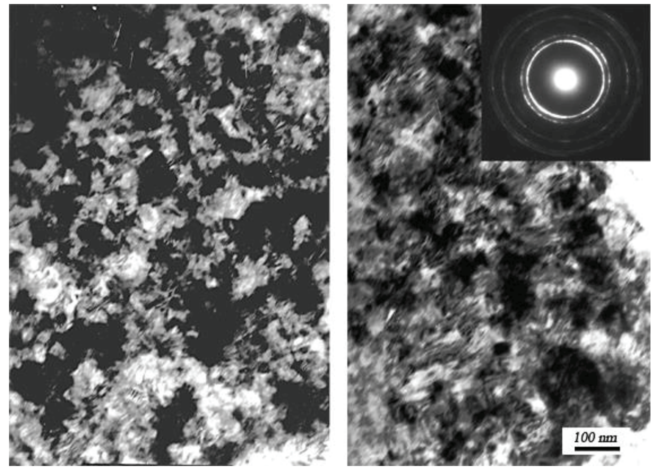


Figure 1: Field dependences of magnetoresistance of $[Co/Cu]_n$ superlattices with n = 10 (a) and n = 40 (b).

The NMR studies were performed in impulse NMR spectrometer at liquid helium temperature (4.2 K) in zero-field, with ^{59}Co as probe-nucleus. The spin-echo signal $E(2t)$ was formed by a sequence of two coherent radio-frequency impulses, $(\tau_p)_x - t_{del} - (\tau_p)_y - t_{del}$ − echo, forming the alternating magnetic field with the circular component amplitude of $H_1 \sim 10$ Oe in the resonance coil containing a specimen. The measurements were performed in the frequency range of 145-235 MHz, with the impulse duration τ_p of 0.5 ms and the time interval between impulses t_{del} of 20 ms. The impulse power was held constant. In the range of 208-235 MHz the measurements were done with a 1 MHz step, and the rest of the spectra were taken with the 2 MHz step. The specimens were 10 × 10 mm^2 plates. The measuring coil and specimen planes were coplanar, the magnetic moment lying in the superlattice plane. All the spectra were taken in similar conditions. Each spectrum was recorded within one measuring session, i.e., without disassembling and reassembling.

3 RESULTS AND DISCUSSION

As demonstrated by TEM, the superlattices under study have nanocrystalline structure, and their crystallites are separated by high-angle boundaries, which is testified by the ring-wise electron diffraction patterns, in which the Debye rings, densely populated by the reflections corresponding to the FCC lattice, are clearly seen (Figure 2). Judging from the electron diffraction patterns, in which all the reflections corresponding to FCC lattices (Cu or Co) are present (see the insert to Figure 2b), it can be concluded that there is no sharp texture in the films under study. Besides, it is evident that the HCP-modification of Co is absent, because there is no line corresponding to the interplanar space of 0.1915 nm, which is the most intensive line (101) of the HCP lattice of Co. According to the bright-field and dark-field images, the crystallites size in the layers plane is about 25 nm. The images look blurred because of the highly dispersed structure.



a b

Figure 2: Electron micrographs of Glass/Fe(5nm)/$[Co(1.5nm)/Cu(0.9nm)]_{40}$/Cr(3nm) multilayer: a - dark field image in the (111) reflections of Cu and Co; b - bright field image and electron diffraction pattern.

Figure 3 demonstrates the results of low-angle X-ray diffraction for metal superlattices $[Co/Cu]_n$, where n = 10, 20, 30 and 40. It is seen that the first Bragg peak for all the nanostructures is close to the angle of 2θ = 4.6°, which means that the superlattice periods are the same in all the specimens. Treating the X-ray reflectometry data we found the minimal root-mean-square deviation of intensity from its theoretical value calculated by the program based on the dynamic theory of X-ray scattering. For the fitting procedure the intensity values measured dependently on the reflection angle up to 2θ = 6° were used. As shown in Figure 3, the Kessing oscillations intensity decreases with the increasing number of bilayers which is due to the increasing number of diffraction planes and decreasing intensity of every peak. On the other hand, the decreasing intensity of Kessing oscillations may be caused by the growing roughness of interlayer interfaces. Numerical treatment also testifies the increasing roughness of interfaces. This assumption will be checked in our further studies.

As demonstrated by the investigation of field dependences of magnetoresistance, the latter decreases with the growing number of bilayers from 38% for n = 10 to 24% for n = 40. The corresponding dependence of the magnetoresistance on the number of bilayers is shown in Figure 4.

Figure 5 demonstrates a typical example of a NMR spectrum. This figure shows a spin echo intensity for the Glass/Fe(5nm)/[Co(1.5nm)/Cu(0.9nm)]40/Cr(3nm) multilayered structure versus frequency, and its expansion into components. We fitted the intensity of spectrums by separate Gaussian lines.

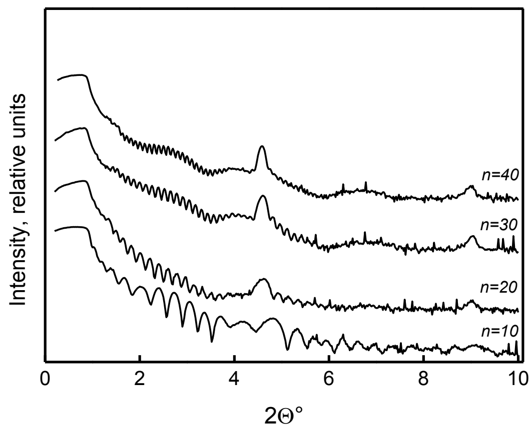


Figure 3: X-ray reflectivity scans of Glass/Fe(5nm)/[Co(1.5nm)/Cu(0.9nm)]$_n$/Cr(3nm) multilayers for n = 10 , 20 , 30 and 40.

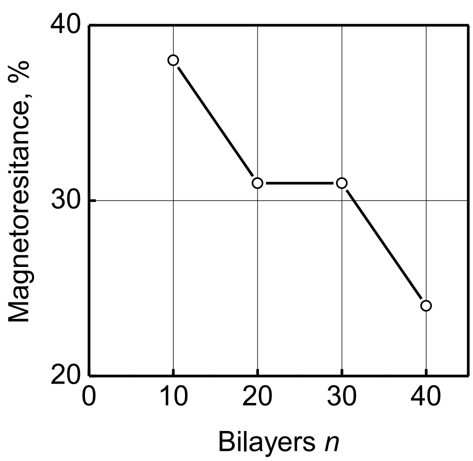


Figure 4: Magnetoresistance versus the number of bilayers.

The spectrums for different numbers of bilayers are qualitatively similar, but the relative intensities of the spectrum components are different. The solid line in Figure 5 shows that the structure of the spectrum is well fitted by four approximately equally spaced Gaussians (denoted by dotted lines). All parameters in the fit were free except for the line width, which was constrained to have the same value for all lines. The main line in the spectra appears close to the value of Co atoms in the FCC lattice surrounded by bulk Co (217 MHz [9]). Consequently, it is formed by Co atoms located in the layers bulk. From the high-frequency side of this line the intensity practically vanishes, which indicates that there are no HCP Co and stacking faults in the specimens under study. The components with resonance frequencies lower than that for the volume line are attributed to Co atoms located at interfaces, where one or more nearest neighbors of Co atoms are substituted for Cu atoms. The average size between the lines is 20 MHz which is close to the value obtained in [9] (20 MHz per Cu atom). Thus, the I_0, I_1, I_2 and I_3 lines correspond to the Co atoms which have 0, 1, 2 or 3 Cu atoms in their nearest neighborhood (and 12, 11, 10 or 9 Co atoms respectively).

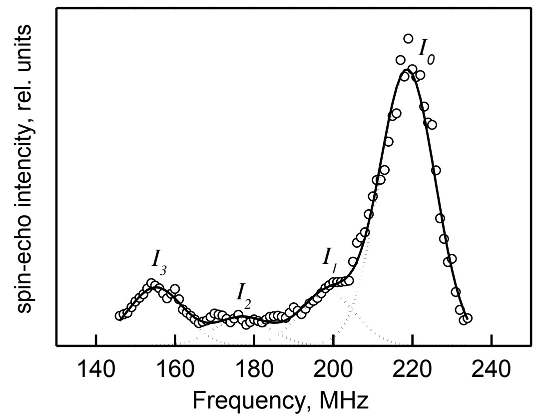


Figure 5. NMR spin-echo intensity versus the frequency for the Glass/Fe(5nm)/[Co(1.5nm)/Cu(0.9nm)]$_{40}$/Cr(3nm) multilayer. Solid line represents the results of fitting by four Gaussians. Each Gaussian (dotted lines) corresponds to Co atoms in specific environment.

In ref. [9] 7 maxima were obtained in the expansion of NMR spectra of analogous superlattices, i.e. the lines formed by Co atoms with the number of the nearest neighbors ranging from 12 to 6 were present. The frequency range of our equipment enables to reveal only 4 peaks. However, this is unlikely to result in introducing an essential error. As stated in [9], the observed distribution of neighbors is not random, as the Co-Cu system tends to disintegration, and thus in an interface layer the Co atoms are preferably surrounded by Co, and Cu atoms by Cu. Thus, the peaks corresponding to a relatively high fraction of Cu in the environment of Co atoms demonstrate weak intensity.

We have analyzed the correlation between the number of bilayers in a superlattice and the interface thickness. The fraction of Co atoms at an interface has been estimated from the ratio of the sum of interface peak intensities to the sum of volume and interface peaks, $(I_1 + I_2 + I_3)/(I_0 + I_1 + I_2 + I_3)$. If an interface average composition is assumed to be equal-atomic and the Co layer thickness is known, one can estimate the interface thickness from the formula $t_{\rm int} = t_{\rm co}(I_1+I_2+I_3)/(I_0+I_1+I_2+I_3)$, where $t_{\rm int}$ is an interface thickness, and $t_{\rm co}$ is a Co layer thickness. Figure 6 demonstrates the as-estimated interface thickness as a function of the number of bilayers. It is seen that with the increase of the bilayers number from 10 to 40 the Co/Cu interface thickness decreases from 0.49 to 0.33 nm.

On the other hand, the internal structure of interfaces can be characterized by the fraction of areas of “perfect” conjunction in the overall surface of interfaces. The I_3 peak characterizes the ^{59}Co atoms of the “ideal” Co/Cu interface coinciding with the closely packed <111> plane of FCC lattice, in which every atom of ^{59}Co has three nearest neighbors of Cu atoms. As mentioned above, we suggest that the contribution of configurations with more than three nearest neighbors may be neglected. Hence, the fraction of the "ideal" interface can be determined as $I_3/(I_1+I_2+I_3)$.

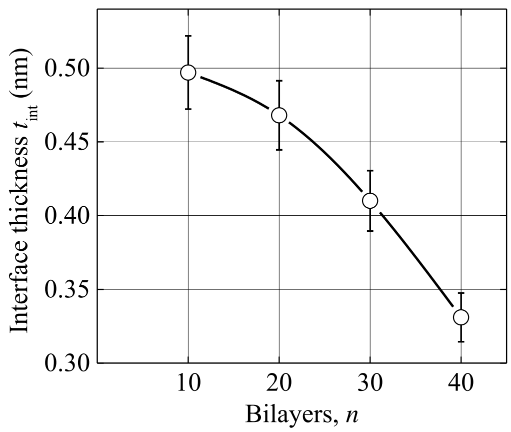


Figure 6: Interface thickness in $[Co/Cu]_n$ superlattices versus the number of bilayers, n.

Figure 7 demonstrates variation of this parameter dependently on the number of bilayers. It is evident, that the fraction of the areas of ideal conjunction decreases with the growth of the number of bilayers.

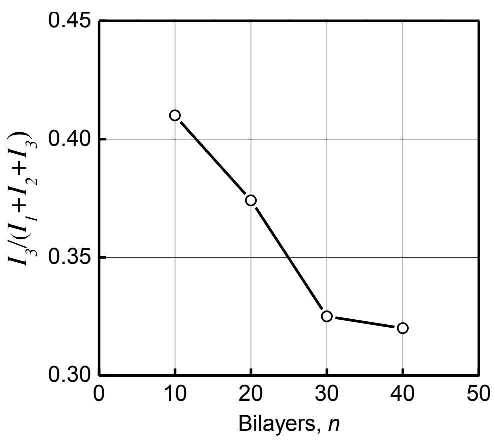


Figure 7: The fraction of Co atoms at "ideal" interfaces versus the number of bilayers, n.

4 SUMMARY

The magnetoresistive properties and structural characteristics of interfaces in multilayers Glass/Fe(5nm)/[Co(1.5nm)/Cu(0.9nm)]$_n$/Cr(3nm) with various number of couples of layers have been studied and analyzed. It has been found that with the increase of the number of bilayers the magnetoresistance decreases. At the same time the interface thickness and the fraction of areas of "ideal" conjunction decrease as well.

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