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Modelling of interface structure in Co/Cu superlattices with Fe buffer layer

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Received October 10, 2024
Revised October 25, 2024
Accepted November 5, 2024

A method of interpreting experimental nuclear magnetic resonance spectra to describe the structure of interlayer boundaries in Co/Cu superlattices with a buffer layer of Fe prepared by magnetron sputtering is proposed. The superlattices have the structural formula glass/Fe(5nm)/[Co(1.5nm)/Cu(1nm)]n/Cr(5nm), $n = 10, 20, 30, 40$. The performed three-dimensional modelling of the structure of interlayer boundaries allows us to make computational NMR spectra that allow us to interpret the experimental spectra. In this work it is demonstrated that in Co/Cu superlattices with the use of Fe buffer layer there is a penetration into copper cobalt at the Co/Cu interface. It is shown that in the interface region „island" copper in cobalt penetrations with a depth of one atomic layer are formed.

Keywords: superlattices, interfaces, NMR, modelling.

DOI: 10.61011/PSS.2024.12.60206.259

1. Introduction

Such type of low-size systems as multilayer nanostructures — superlattices - are currently used in the development of spintronic and electronic devices, magnetic field sensors [1–4]. Interest in superlattices is driven by the giant magnetoresistance effect [5,6] that is observed in them. Superlattices have an essential property — magnetoresistance that in turn depends on a set of parameters: substrate and buffer layer material [7–11], number of pairs of layers [12,13], magnetic and non-magnetic layer thicknesses [7,14,15], heat treatment conditions [16,17], preparation procedure and conditions [18–20]. The above-listed parameters define the state of magnetic and non-magnetic layer interfaces. State (structure) of interfaces is the factor affecting the spin transport behavior in superlattices [21–23]. The following methods are used to determine the structure of layer interfaces: transmission electron microscopy [24], X-ray diffraction [25], X-ray reflectometry [26], EXAFS [27], Mossbauer spectroscopy [28], nuclear magnetic resonance (NMR) [29]. Previously, the author used X-ray diffraction, X-ray reflectometry and nuclear magnetic resonance to determine the state of interfaces in Co/Cu superlattices and to identify how the state of these interfaces affects the magnetoresistance [30]. The study has identified how the state of interfaces affect the magnetoresistance through interpreting the experimental NMR spectra. However, the study mentioned above estimated the number of seamless interfaces and showed that the interfaces have roughness, however, numerical estimate of roughness is not provided.

There are currently numerous publications devoted to determining interface structure according to the nuclear magnetic resonance data [12,31]. There are also publications offering various interface structure models that were used to get calculated NMR spectra [7,14]. Thus, in [14] it is shown that, in the [Co(1.23nm)/Cu(4.2nm)] superlattices prepared on the SiO_2 substrate and with Cu buffer layer, many insertions with a depth of one atomic layer are formed at the magnetic and non-magnetic layer interface, and Co and Cu atom distribution in the interface area is not occasional. In [7], a two-dimensional interface model from the study mentioned above is supplemented and developed. This model admitted large interface thickness and also accounted for atom concentration depending on the interface thickness. Authors of [7] suggest that locally more flattened interfaces are formed in the studied superlattices, and transition to the island model will help achieve better description of experimental data using the calculated nuclear magnetic resonance spectra. In [10], a modified model from [14] is also used. Using interface structure modeling, the author has estimated the influence of the Fe buffer layer thickness on the interface thickness.

The provided models make it possible to interpret experimental NMR data for interface structure description. Note that these models are mainly restricted to the two-dimensional interface representation. Therefore, transition to three-dimensional models is important and will help get new information on the interface structure. This study provides three-dimensional modeling of the interface structure in Co/Cu superlattices with Fe buffer layer. Using the three-dimensional models, NMR spectra were calculated to interpret the experimental nuclear magnetic resonance

data used to get new information regarding the features of the layer interface structure in the Co/Cu cobalt-containing superlattices.

2. Samples and experimental procedure

The studied superlattices were prepared by the magnetron sputtering method. Structural formula of superlattices is: glass/Fe(5nm)/[Co(1.5nm)/Cu(1nm)]$_n$/Cr(5nm), $n = 10$, 20, 30, 40.

X-ray diffraction patterns were recorded using PANalytical Empyrean Series 2 software and hardware suite in CoK_α radiation.

The nuclear magnetic resonance spectra on ^{59}Co nuclei were recorded without an external magnetic field at liquid helium temperature of 4.2 K. Upgraded „Bruker" phase-coherent spectrometer was used for recording. Spectra were recorded by frequency scanning method in the frequency range of 235−110 MHz. The spin echo signal was formed by a sequence of two coherent radio frequency (RF) pulses — a solid echo pulse sequence. Pulse duration $\tau_{\text{pulse}} = 0.5\,\mu$s, pulse-to-pulse spacing $\tau_{\text{delay}} = 11\,\mu$s. Distance between the experimental points 1 MHz. To remove distortion of the spectra due to the transient processes and interference effects in the resonance circuit, a RF pulse phase alternation sequence was used. To increase the signal-to-noise ratio, multiple signal integration was carried out. Amplifier power constancy was monitored throughout the operating range.

As long as magnetic moments of the Co layers are oriented along the geometrical plane of the film, so a standard solenoid may not be used as a test coil (TC) because the magnetic induction vector is co-directional with the superlattice magnetization vector. In [32], it was shown that the optimum shape of TC — is a flat helical coil. To observe the NMR signal, TC was made (Figure 1).

Test coil — is a flat wire helix 113 mm in length and 0.43 mm in diameter (varnish) with soldered central tap contact. The structure is placed in a plastic housing filled with epoxy. To achieve maximum Q factor of the resonant circuit, thin mica wafers between the sample and TC were used.

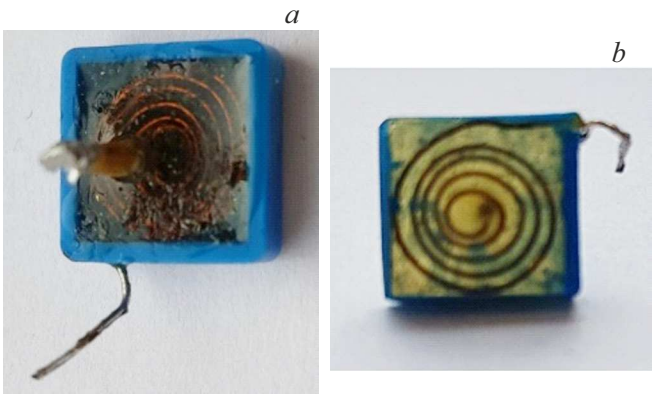

Figure 1. Test coil for Co/Cu superlattices: *a* — top view; *b* — bottom view.

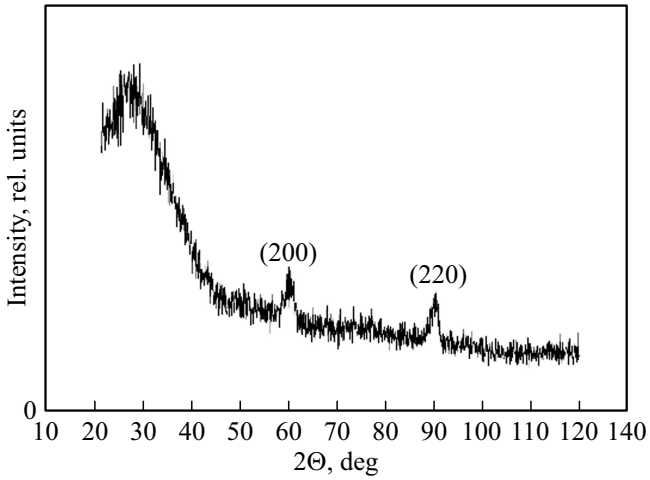

Figure 2. X-ray diffraction pattern of the glass//Fe(5nm)/[Co(1.5nm)/Cu(0.9nm)]$_{30}$/Cr(3nm) superlattice, radiation CoK_α, $\lambda = 1.7889$ Å.

3. Findings and discussion

The type of crystal structure and texture formed in superlattices defines the structure of seamless interfaces. Figure 2 shows an X-ray diffraction pattern of the glass//Fe(5nm)/[Co(1.5nm)/Cu(1nm)]$_{30}$/Cr(5nm) superlattice.

In the range of $20° \le \Theta \le 120°$, two Bragg peaks were found that refer to the (200), (220) reflexes that are common to Co and Cu, which is indicative of a two-component texture in the studied superlattices. X-ray reflectometer method confirmed the existence of a periodic structure, the superlattice period is equal to the nominal period (2.4 nm), slow reduction of the Kissig oscillation amplitude is indicative of high quality of interfaces. Interpretation of reflectograms has shown that roughness was not higher than 0.4 nm (for the glass//Fe(5nm)/[Co(1.5nm)/Cu(0.9nm)]10/Cr(3nm) superlattice). A face-centered cubic (FCC) type crystal structure was formed in the superlattices. Figure 3, *a* shows a seamless layer interface for the (100) texture, Figure 3, *b* shows a seamless layer interface for the (110) texture.

In Figure 3, Co atoms are dark grey, Co atoms are black. Black lines interconnect the selected Co atom with Cu atoms in its immediate environment. Figure 3, *a* shows that, for the (100) texture, the selected Co atom involved in formation of the seamless layer interface has 4 Cu atoms in the immediate environment, and for the (110) texture — the selected Co atom has 5 Cu atoms in the immediate environment.

Let's consider how the nuclear magnetic resonance method is used to get interface structure data. Due to

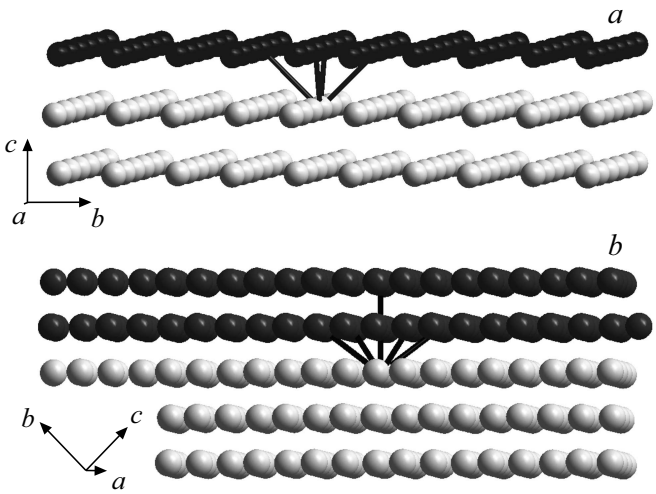

Figure 3. Three-dimensional model of a seamless interface in the glass//Fe(5nm)/[Co(1.5nm)/Cu(0.9nm)]$_{30}$/Cr(3nm) superlattices for the (100) (*a*) and (110) (*b*) textures.

hyperfine interaction, magnetic moments of the Co atoms induce local magnetic fields in the ^{59}Co nucleus locations; strength and direction of the magnetic fields depend on the magnetic and structural features of the immediate environment of the probe nucleus. In [33], the value of the hyperfine filed was measured experimentally: — 21.6 T. Substitution of one Co atom with one Cu atom in the immediate environment of the probe nucleus leads to reduction of the hyperfine filed by 1.6–1.8 T [34,35]. Also in [33], expression (1) is provided to estimate the hyperfine field depending on the number of substituted Co atoms in the immediate environment of the probe nucleus:

$$H_{hf} \approx H_{hf}^{b} - \Delta H_{hf}^{1}(n^{b} - n^{1}), \qquad (1)$$

where H_{hf}^{b} is the hyperfine field in a bulk material, n^{b} is the coordination number in a bulk material, shift ΔH_{hf}^{1} is about -1.8 – -1.6 T [14], n^{1} is the number of Co atoms in the first coordination sphere for the given Co probe nucleus. This expression means the following: if the first coordination sphere of the Co atom contains 12 Co atoms (because a FCC structure was formed in the studied superlattices), then the resonance frequency (including the gyromagnetic ratio) of the probe nucleus will be 216 MHz. Substitution of a Co atom with a Co atom in the immediate environment of the probe nucleus will lead to reduction of the resonance frequency to 200 MHz. Substitution of two Co atoms with two Cu atoms will lead to formation of a resonant line at 184 MHz, etc. According to Figure 3, in the case of the (100) texture, the resonance line corresponding to Co atoms, that form seamless layer interfaces, is observed at 150 MHz, and in the case of the (110) texture, the resonance line of Co atoms, that form the seamless interfaces, is observed at 132 MHz. In the case of the (111) texture and when only seamless layer interfaces exist in the superlattice, the experimental NMR spectrum has only two lines: at 216 MHz —Co atoms within the Co layer, and at 168 MHz — the resonance line characterizing the Co atoms involved in formation of the layer interfaces. Such experimental spectrum is described in [36], where the Co/Cu superlattices were grown by the molecular-beam epitaxy method. Figure 4 shows the nuclear magnetic resonance spectrum of the glass//Fe(5nm)/[Co(1.5nm)/Cu(0.9nm)]$_{10}$/Cr(3nm) superlattice:

Figure 4 shows position of the resonance lines I_j, where j — is the number of Co atoms substituted by the Cu atoms in the immediate environment of the probe nucleus. The shape of the line is described using the Gaussian function and the line width is the same for all resonance lines. Line width was determined during interpreting the NMR spectra. The presence in the NMR spectrum of the resonance lines I_0 (Co atoms within the Co layer), I_4 (Co atoms forming the seamless layer interfaces for the (100) texture) and I_5 (Co atoms forming the seamless layer interfaces for the (110) texture) implies non-zero roughness of Co/Cu layer interfaces.

Three-dimensional modeling of the layer interface structure and estimation of the interface roughness were performed in accordance with the trend of Cu penetration into Co at the Co/Cu interface [26]. Modeling takes into account the assumption from [7] that locally more flattened interfaces than those implied in the superlattice models may be implemented in the superlattices, and the Cu insertions into Co may have an „island“ shape. Various types of Cu insertion into Co were used for modeling: Cu atom chain penetrating Co to a depth of one atomic layer, insertions in the form of needle and cone with different depths. However, the listed-above Cu insertions into Co in the interface area prevented from achieving a satisfactory coincidence between the calculated and experimental spectra. Figure 5 shows three-dimensional models of the Co/Cu interface structure for the (100) texture (Figure 5, *a*)

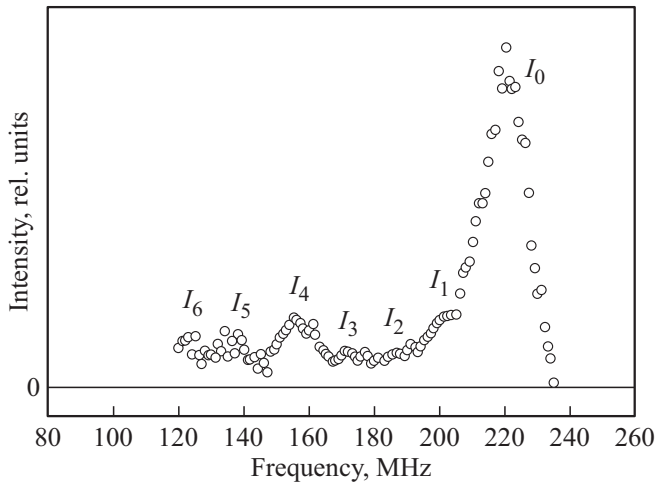

Figure 4. Experimental NMR spectrum of the glass//Fe(5nm)/[Co(1.5nm)/Cu(0.9nm)]$_{10}$/Cr(3nm) superlattice.

and (110) texture (Figure 5, *b*), and the calculated and experimental nuclear magnetic resonance spectra for the (100) texture (Figure 5, *c*) and (110) texture (Figure 5, *d*) in the Co/Cu superlattices.

Figure 5, *c, d* shows a high degree of coincidence between the experimental and calculated spectra. As long as a two-component texture is formed in these superlattices, then the resulting calculated spectra shall be summarized (Figure 6).

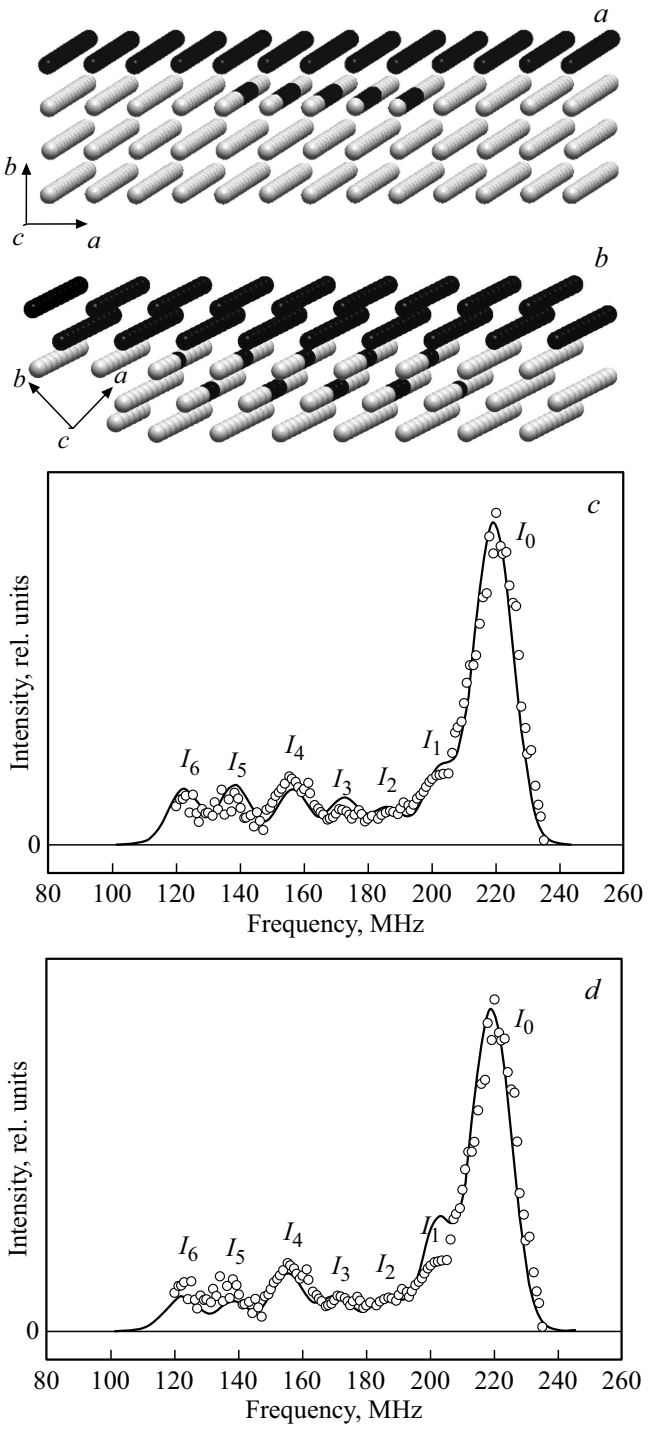

Figure 5. Interface structure in the case of the (100) (*a*), (110) (*b*) textures and corresponding calculated NMR spectra (*c, d*). (*c, d*) show experimental NMR spectrum of the glass//Fe(5nm)/[Co(1.5nm)/Cu(0.9nm)]$_{10}$/Cr(3nm) superlattice.

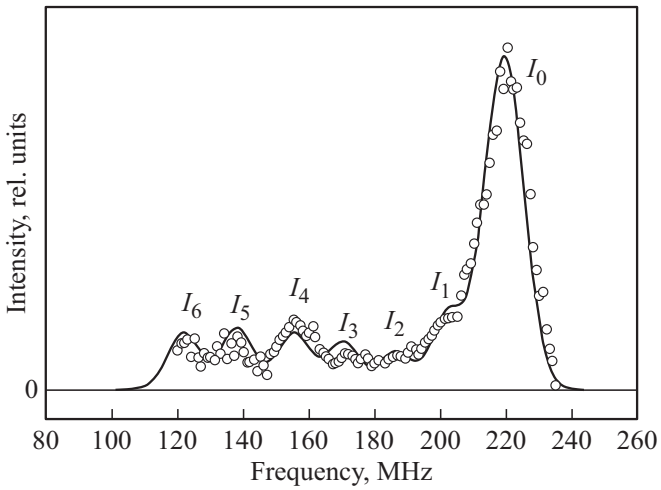

Figure 6. Calculated and experimental NMR spectra of the glass//Fe(5nm)/[Co(1.5nm)/Cu(0.9nm)]$_{10}$/Cr(3nm) superlattice.

Figure 6 shows that the calculated NMR spectrum obtained on the basis of the proposed „island" model of Cu insertion into Co in the interface area allows description of the experimental NMR spectrum.

4. Conclusion

The experimental nuclear magnetic resonance data was used to determine local field distribution in the glass//Fe(5nm)/[Co(1.5nm)/Cu(1nm)]$_n$/Cr(5nm) superlattices, $n = 10$, 20, 30, 40 prepared by the magnetron sputtering method. The X-ray diffraction method identified that a two-component (100), (110) texture had been formed in these superlattices. Three-dimensional modeling of the interface structure was performed, „island" model of Cu insertion into Co was proposed. Three-dimensional modeling data was used to calculate the NMR spectrum allowing description of the experimental NMR spectra. Thus, it is shown that „island" insertions of Cu into Co with a depth equal to one atomic layer are formed.

Acknowledgments

The author is grateful to M.A. Milyaev for the provided superlattices. X-ray examinations were conducted using the equipment provided by the Shared Research Facility of the Nanospintronics Department, Institute of Metal Physics, Ural Branch, Russian Academy of Sciences.

Funding

The study was performed under state assignment No. 122021000035-6 on topic „Function".

Conflict of interest

The author declares no conflict of interest.

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Translated by E.Ilinskaya