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MAGNETISM AND FERROELECTRICITY

Ferromagnetic and Spin-Wave Resonance in Co/Pd/CoNi Multilayer Films

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Abstract—It has been found that the magnitude and sign of exchange interaction between Co(5 nm) and CoNi(5 nm) ferromagnetic layers through Pd depend on magnetization orientation of ferromagnetic layers. If magnetization is oriented in a layer plane, exchange interaction can be both ferromagnetic and antiferromagnetic. If magnetization orientation is orthogonal to a layer plane, the exchange constant is always positive at $d_{\mathrm{Pd}} < d_c$ and equals zero at $d_{\mathrm{Pd}} > d_c$ (d_c is the characteristic length). *© 2003 MAIK "Nauka/Interperiodica".*

1. INTRODUCTION

A great number of experimental and theoretical papers [1, 2] are devoted to investigation of the magnetic properties of multilayer systems with alternative layers of ferromagnet/ferromagnet, ferromagnet/paramagnet, ferromagnet/diamagnet types. The interest in these systems is explained by the possibility of creating film materials with a set of necessary magnetic properties by selecting the thickness and chemical composition of individual layers. It is well known that ferromagnetic layers formed of Fe, Co, Ni, and their alloys and separated by nonferromagnetic layers of Ag, Pt, Cr, Cu, etc., can be exchange-coupled either ferromagnetically or antiferromagnetically depending on the nonferromagnetic layer thickness. This exchange interaction between ferromagnetic layers separated by a metal interlayer leads to the formation of a magnetically unified system in a multilayer film. As a result, the integral electrical and magnetic characteristics (remagnetization curve, perpendicular anisotropy, etc.) of these multilayer structures are mostly governed by some effective exchange interaction. At the same time, traditional methods (low-temperature run of saturation magnetization, neutron diffraction analysis, etc.) used to evaluate the exchange integral describing the interaction of a local magnetic moment with its nearest surroundings in ferromagnets turn out to be poorly informative for the study of effective exchange in multilayer structures. Resonance methods of investigation, i.e., ferromagnetic resonance (FMR) and spin-wave resonance (SWR), seem to be the only methods which allow both the detection of signals caused by this exchange-coupling effective constant and measurement of the value of this constant in such composite materials as ferromagnetic multilayer films.

In this paper, we report the results of an experimental study on the FMR and SWR spectra in a complex composition system, Co/Pd/CoNi multilayer films, with the aim of determining the effective exchange-coupling constant and of finding the dependence of this constant on the thickness of individual layers.

2. EXPERIMENTAL

The main attention in the work was paid to the study of the resonance characteristics of Co/Pd, Co/CoNi, and Co/Pd/CoNi multilayer films in two different orientations (external field parallel or perpendicular to the film plane). Experimental results allowed us to determine the exchange-coupling effective constants of these composite materials and to calculate partial values of the exchange-coupling constant in individual layers of a multilayer film.

SWR is known to be a very sensitive and specific method of investigation with certain requirements on the quality of the film (absence of macroscopic heterophase zones in the layer plane, narrow resonance-absorption line, etc.). We used a chemical precipitation method which allowed us to synthesize high-quality multilayer structures suitable for resonance investigation.

Let us consider FMR and SWR spectra of Co/Pd and Co/Pd/CoNi multilayer films. SWR spectra of these films were studied using a standard x-band spectrometer with a frequency of the resonator pumping $f = 9.2$ GHz at room temperature. For ferromagnetic films 200 nm thick, SWR is usually realized in the range of wavenumbers of standing spin waves from 10^5 to 10^6 cm^{-1}.

Multilayer films for the investigation were obtained through chemical precipitation from a water solution of

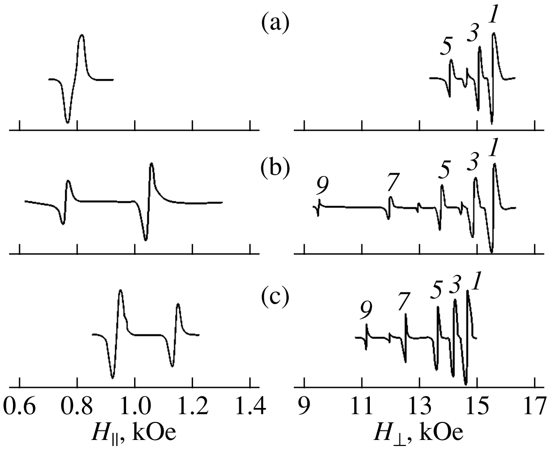

Fig. 1. Typical resonance spectra of three series of samples measured in two orientations: magnetic field ($H_{||}$) parallel and ($H_{\perp}$) perpendicular to the film plane. (a) Co/Pd series; (b) Co/Pd/CoNi series; and (c) Co/CoNi series.

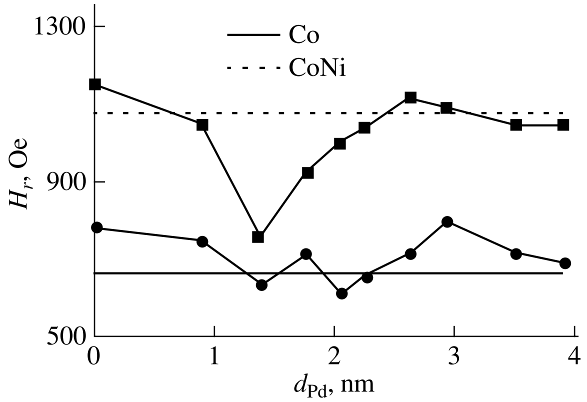

Fig. 2. The dependence of the ferromagnetic resonance fields of individual ferromagnetic layers on Pd-interlayer thickness in Co/Pd/CoNi films for samples oriented parallel to an external magnetic field.

the appropriate metal salts onto a glass substrate. The first series of samples consisted of three pairs of Co/Pd layers. The Co layer thickness $d_1 = 5$ nm was constant, and the Pd-layer thickness d_2 varied from 0.5 to 4 nm; the total thickness of the film did not exceed 30 nm. Previous x-ray study of Co–P thin films showed that, if 5–9 at. % P is introduced into a Co solution, the precipitate has the structure characteristic of a face-centered cubic (fcc) packing of atoms. Thus, the introduction of 7 at. % phosphorus provided fcc-structure in the Co layers in CoPd multilayer films and undistorted fcc-structure in the Pd layers precipitated on Co layers. The second series of samples under investigation differed from the first one in the introduction of an additional CoNi layer to the Co/Pd multilayer structure. The presence of 20 at. % Ni in the Co solution allowed us to form ferromagnetic layers with different magnetization in [Co/Pd/CoNi] $*$ 7 multilayer films. As in the first series of samples, the Co and CoNi ferromagnetic layer thicknesses were constant, $d_1 = 5$ nm, while the Pd-layer thickness d_2 varied from 0.5 to 4 nm; the total thickness of the [Co(5 nm)/Pd(x)/CoNi(5 nm)] $*$ 7 film varied from 70 to 130 nm. The third series of multilayer films was produced as [Co(X nm)/CoNi(X nm)] $*$ 10 type. The thicknesses of the Co and CoNi ferromagnetic layers were selected equal and varied from 2 to 10 nm, so that the total thickness of these films varied from 40 to 200 nm.

The resonance characteristics of these three series of samples, measured in two orientations (magnetic field parallel or perpendicular to the film surface), are shown in Fig. 1. As is evident from the experimental curves, the FMR spectrum in parallel geometry in Co/Pd samples is represented by a single resonance line, the coordinate (resonance field H_r value) and width ΔH_r of which weakly depend on Pd-layer thickness. This resonance field magnitude can be determined from a standard expression:

$$\frac{\omega}{\gamma} = [H(4\pi M_{\mathrm{eff}} + H)]^{1/2}, \qquad (1)$$

where γ is the gyromagnetic ratio.

The films of the second and third series featured a specific FMR spectrum; at parallel orientation of both [Co(5 nm)/Pd(x)/CoNi(5 nm)] $*$ 7 and [Co/CoNi] $*$ 10 films to the external field, there were two distinct resonance peaks in the FMR spectrum, i.e., low-field (caused by Co layers) and high-field peaks (caused by CoNi layers). The dependences of resonance fields of individual ferromagnetic layers on Pd-layer thickness in Co/Pd/CoNi films are shown in Fig. 2. These results cannot be described using formula (1); therefore, it requires modification. When the film is oriented perpendicular to a magnetic field, the SWR spectrum is observed in all three series. SWR spectra in Co/Pd/CoNi and Co/CoNi films have as many as nine peaks and in Co/Pd films up to five peaks. The analysis of the spectra allows us to conclude that surface spin pinning similar to Kittel pinning takes place here; the intensities of odd peaks exceed those of even peaks by more than an order of magnitude. The angular dependence, peak-intensity behavior and the arrangement of the peaks indicate that spin-wave modes are observed in these films. Resonance fields of these modes are described by a standard Kittel expression:

$$\frac{\omega}{\gamma} = H - 4\pi M_{\mathrm{eff}} + \frac{2A_{\mathrm{eff}}}{M}\left(\frac{n\pi}{d}\right)^2, \qquad (2)$$

where n is a peak number, d is the total film thickness, and M_{eff} and A_{eff} are the basic parameters of the composite system requiring determination. By plotting H_r vs. the square of n, we can measure the value of A_{eff} experimentally. Dependences of the resonance field H_r

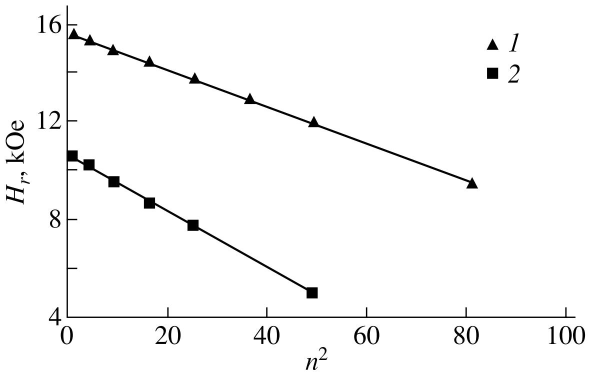

Fig. 3. Typical dependences of resonance field H_r on the square of the spin-wave mode index n of SWR spectra for Co/Pd/CoNi films. (*1*) [Co(5 nm)/Pd(0.9 nm)/CoNi(5 nm)] ∗ 7; and (*2*) [Co(5 nm)/Pd(2.7 nm)/CoNi(5 nm)] ∗ 7.

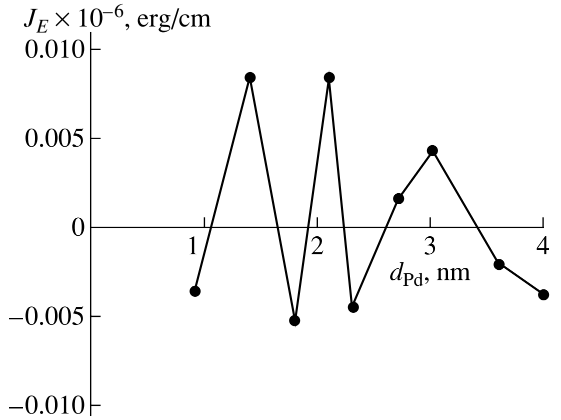

Fig. 4. The dependence of the constant of interlayer exchange interaction in Co/Pd/CoNi films on Pd-layer thickness as calculated from FMR spectra obtained at parallel orientation of the films in an external field.

on the square of the spin-wave mode n^2 of the SWR spectra in the [Co(5 nm)/Pd(x)/CoNi(5 nm)] ∗ 7 multilayer film series are shown in Fig. 3 (the SWR spectrum shape is shown in Fig. 1).

3. ANALYSIS OF EXPERIMENTAL RESULTS

Let us assume that the shift in the CoNi- and Co-layer resonance peaks in Co/Pd/CoNi multilayer films from the H_r values for reference films (Co and CoNi single-layer films were characterized by a single resonance peak corresponding to the conventional FMR) results from exchange modification of the value of the internal field in individual layers [3]. An expression for this exchange modification can be obtained using a model of a connected two-layer film system consisting of two ferromagnetic layers interacting through an intermediate nonmagnetic layer with interaction energy per unit area $E_{AB} = -J_E\mathbf{M}_A\mathbf{M}_B$, where J_E is the exchange-interaction constant and the vectors $\mathbf{M}_A$ and $\mathbf{M}_B$ denote magnetization in layers A and B, respectively [4]. If $d_A = d_B = d$ and the anisotropy field can be ignored ($H_{\mathrm{Keff}A} = H_{\mathrm{Keff}B} = 0$), we assume that it is possible to express the total energy for a two-layer system of interaction in the following form:

$$E = [-(\mathbf{HM}_A) + 2\pi M_A^2\cos^2\Theta_A]d + [-(\mathbf{HM}_B) + 2\pi M_B^2\cos^2\Theta_B]d - J_E\mathbf{M}_A\mathbf{M}_B. \tag{3}$$

The dispersion relation for this system is defined by the solution to the Landau–Lifshitz equation and can be written as

$$\begin{aligned}&(\omega/\gamma)^4 - (\omega/\gamma)^2[H4\pi(M_A + M_B)\\&+ 8\pi J_E M_A M_B + J_E^2(M_A + M_B)^2] + H^2 16\pi^2 M_A M_B\\&+ H16\pi^2 M_A M_B J_E(M_A + M_B)\\&+ H4\pi J_E^2(M_A + M_B)(M_A^2 + M_B^2) = 0.\end{aligned} \tag{4}$$

At large values of J_E, this relation has roots corresponding to acoustical and optical oscillations of magnetization vectors in the A and B ferromagnetic layers. If $J_E \longrightarrow 0$, there are also two roots which describe two resonance curves for zones with different magnetization values (M_A, M_B):

$$\begin{aligned}\left(\frac{\omega^2}{\gamma^2}\right)_1 &= 4\pi M_A H + J_E M_B M_A,\\ \left(\frac{\omega^2}{\gamma^2}\right)_2 &= 4\pi M_B H + J_E M_A M_B.\end{aligned} \tag{5}$$

Plotting the experimental values of the resonance field for the two modes obtained from FMR spectra for the Co/Pd/CoNi multilayer film and fitting the curves calculated from equation (5) allows us to determine the dependence $J_E(d_{\mathrm{Pd}})$ (see Fig. 4). The calculated $J_E(d_{\mathrm{Pd}})$ magnitudes vary in the range from 0.005 to 0.01 erg/cm depending on the Pd-layer thickness. The oscillation period $J_E(d_{\mathrm{Pd}})$ is equal to 0.7 nm. It can be seen that the $J_E(d_{\mathrm{Pd}})$ dependence is described by the product of an oscillating function and some function f, which decreases with increasing d_{Pd}.

Our previous systematic investigation of the magnetooptical properties (Faraday effect, equatorial Kerr effect) of Co/Pd and Co/Pd/CoNi multilayer films [5, 6] revealed corresponding oscillations, which allows us to conclude that the electronic structure of these composite materials is different from that of Co single-layer films. Thus, analysis of the specific features in the FMR spectrum for Co/Pd/CoNi films, as well as of the specific features of magnetization curves for Co/Pd films, shows that individual ferromagnetic layers in these films are linked to each other through an interlayer exchange interaction J_E, the magnitude and sign of which change as the Pd-layer thickness varies.

We now consider the SWR spectra. Plotting the linear dependence $H_r(n^2)$ allowed us to calculate the exchange-coupling effective constant A_{eff} for the composite material formed by combinations of Co/Pd, Co/Pd/CoNi, and Co/CoNi multilayer films. For example, it can be seen from dependence $A_{\text{eff}}(d_{\text{Pd}})$ for Co/Pd/CoNi films in Fig. 5 that, while d_{Pd} changes from 1 to 3 nm, the value of A_{eff} increases monotonically; at $d_{\text{Pd}} > 3$ nm, it is evidently equal to zero. The latter fact means that there are no standing spin waves which propagate through the entire sample thickness in Co/Pd/CoNi multilayer films with Pd-layer thicknesses exceeding 3 nm; i.e., $A_{\text{eff}}(d_{\text{Pd}} > d_c) = 0$. An important point is that the experimental $A_{\text{eff}}(d_{\text{Pd}})$ values for the Co/Pd/CoNi system are considerably smaller than the reference A_{eff} values calculated for Co and CoNi ferromagnetic films. Co and CoNi single-layer 200-nm-thick films were chosen as reference samples. The SWR spectra were measured, the $H_r(n^2)$ dependences were constructed, and then the A values were measured for reference films at perpendicular film orientation in an external magnetic field. The obtained reference values $A_{\text{Co}} = 1.2 \times 10^{-6}$ erg/cm and $A_{\text{CoNi}} = 0.5 \times 10^{-6}$ erg/cm coincide with the known tabulated values for similar film samples.

It should be noted that, according to the basic theory on employing the SWR method for multilayer structures, the exchange effective constant determined from SWR spectra for the multilayer structure under investigation (Co/Pd/CoNi) depends on both the partial exchange-coupling constant A_i, characteristic of Co and CoNi layers and the partial exchange between ferromagnetic layers that takes place through Pd layers. The following expression for determining exchange-coupling effective constant of a multilayer film in terms of partial exchanges in individual ferromagnetic metal layers, which form the multilayer system, was suggested in [7]:

$$\frac{d}{A_{\text{eff}}} = \frac{d_1}{A_1} + \frac{d_2}{A_2} + \dots. \qquad (6)$$

Here, d_1 and d_2 are the thickness of different-composition individual layers in the multilayer films with A_1 and A_2 exchange constants, respectively, and $d = d_1 + d_2$ is the multilayer-film period. It is easy to show that, if the condition $d_1 = d_2$ is satisfied, expression (6) for A_{eff} is equivalent [8] to

$$A_{\text{eff}} = \langle A\rangle[1-(\Delta A/\langle A\rangle)^2], \qquad (7)$$

where $\langle A\rangle = (A_1 + A_2)/2$, $\Delta A = A_1 - \langle A\rangle = \langle A\rangle - A_2 \equiv (A_1 - A_2)/2$.

Thus, expression (7) can be used to calculate partial exchanges for Co/CoNi films produced with equal thickness of the ferromagnetic layers.

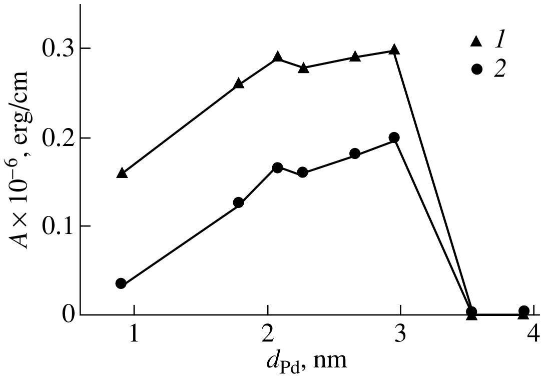

Fig. 5. (*1*) The dependence of the exchange constant obtained from SWR spectra at perpendicular orientation of Co/Pd/CoNi film samples in an external magnetic field on Pd-layer thickness, and (*2*) calculated dependence for the constant of exchange interaction through Pd layers A_{Pd}.

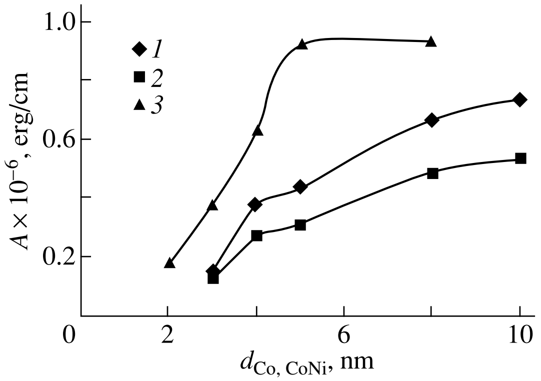

Fig. 6. The dependence of the exchange-coupling constant on Co and CoNi ferromagnetic-layer thickness: (*1*) results obtained from SWR spectra for Co/CoNi films; (*2*) magnitude of the exchange constant in CoNi layers of Co/CoNi films calculated using (8); and (*3*) the exchange constant obtained from the Bloch law ($T^{3/2}$) for [Co(x)/Pd(1.4 nm)] ∗ 25 samples.

The presence of up to nine peaks described by expression (2) in SWR spectra for these films and the linear dependence $H_r(n^2)$ allowed us to calculate the exchange-coupling effective constant A_{eff} for this [Co(x)/CoNi(x)] ∗ 10 system (curve *1*, Fig. 6). Taking into account that the thickness of the ferromagnetic layers for these multilayer films are equal, we used (6) and (7) to derive the following expression for the partial-exchange constant for CoNi:

$$A_{\text{CoNi}} = \frac{A_{\text{eff}}(\alpha + 1)}{2}. \qquad (8)$$

Here, the exchange-constant ratio $\alpha = A^c_{\text{CoNi}}/A^c_{\text{Co}}$ can be replaced by the reference-constant ratio obtained from SWR spectra for CoNi and Co reference single-layer films 200-nm thick. The obtained dependence $A_{\text{CoNi}}(d_{\text{Co, CoNi}})$ is shown in Fig. 6 (curve *2*).

It can be seen from the dependences shown in Fig. 6 that the effective- and partial-exchange constants in Co/CoNi films decrease drastically as the ferromagnetic-layer thickness decreases. For the samples with a ferromagnetic-layer thickness smaller than 5 nm, this value is smaller than the reference values obtained for Co and CoNi single-layer films. Therefore, for Co and CoNi layers each 5-nm thick, each partial-exchange constant should be assumed to be equal to 0.63×10^{-6} and 0.31×10^{-6} erg/cm, respectively. It should be noted that the exchange constants calculated from thermomagnetic curves (Bloch law) for Co/Pd films are consistent with the analogous values of A_{Co} and A_{CoNi} calculated from SWR spectra (curve *3*, Fig. 6).

Thus, having determined the partial-exchange constants in 5-nm-thick Co and CoNi layers and having rewritten expression (6) for the case Co/Pd/CoNi as

$$\frac{d_{\mathrm{Co}} + d_{\mathrm{Pd}} + d_{\mathrm{CoNi}}}{A_{\mathrm{eff}}} = \frac{d_{\mathrm{Co}}}{A_{\mathrm{Co}}} + \frac{d_{\mathrm{Pd}}}{A_{\mathrm{Pd}}} + \frac{d_{\mathrm{CoNi}}}{A_{\mathrm{CoNi}}}, \tag{9}$$

we can use experimental values of A_{eff} to calculate A_{Pd}, the constant of exchange coupling through Pd layers in [Co(5 nm)/Pd(x)/CoNi(5 nm)] $*$ 7 multilayer films. The calculations of A_{Pd} were carried out using the formula

$$A_{\mathrm{Pd}} = \frac{A_{\mathrm{eff}} d_{\mathrm{Pd}}}{(2d_{\mathrm{Co,\,CoNi}} + d_{\mathrm{Pd}}) - A_{\mathrm{eff}} d_{\mathrm{Co,\,CoNi}}(1/A_{\mathrm{Co}} + 1/A_{\mathrm{CoNi}})}, \tag{10}$$

where $d_{\mathrm{Co,\,CoNi}}$ and d_{Pd} are the ferromagnetic and palladium layer thickness, respectively, and A_{Co} and A_{CoNi} are the partial-exchange-coupling constants in the Co and CoNi layers.

The results of calculating, in this manner, the partial constant of exchange interaction through a Pd layer in relation to interlayer thickness for Co/Pd/CoNi multilayer films are also shown in Fig. 5. It can be seen that the calculated $A_{\mathrm{Pd}}(d_{\mathrm{Pd}})$ values are two to three times smaller than the A_{eff} values. However, the functional dependence $A_{\mathrm{Pd}}(d_{\mathrm{Pd}})$ turned out to be similar to the $A_{\mathrm{eff}}(d_{\mathrm{Pd}})$ dependence. The data shown in Fig. 5 suggest that $A_{\mathrm{Pd}}(d_{\mathrm{Pd}})$ is always positive and increases gradually as the palladium interlayer thickness increases up to a critical thickness d_c.

The results of our experiments show that the model usually used to describe ferromagnetic-layer interaction through a nonmagnetic metal layer and to define this interaction in terms of a quasi-Heisenberg isotropic Hamiltonian $J_{12}(d_y)\mathbf{M}_1 \cdot \mathbf{M}_2$ needs to be modified. The simplest modification consists in the following. It is known that an anisotropic Heisenberg Hamiltonian can be written as

$$H = -2\sum_{j>i}\sum [J_z S_i^z S_j^z + J_\perp (S_i^x S_j^x + S_i^y S_j^y)], \tag{11}$$

where S_i and S_j are the spins of neighboring atoms (the superscripts denote the spin-function components along the corresponding axes of the Cartesian system), J_z is the component of exchange-coupling constant along the z axis, and $J_\perp$ is the analogous component in the xy plane. Therefore, if the plane of the multilayer film under investigation coincides with the xy plane of a chosen coordinate system, only the $J_\perp$ component of the exchange-coupling constant can be determined from measuring FMR spectra at parallel orientation of the sample relative to an external magnetic field. In this case, it is reasonable to assume that SWR investigation yields information on the z component of the aforementioned quantity.

Thus, according to the above assumptions, it follows from our experiments that, in Co/Pd/CoNi multilayer films, the exchange constant component in the film plane (xy) features oscillating behavior with increasing palladium-layer thickness (FMR measurements), while the z component of the exchange constant (calculated from SWR spectra) gradually increases with increasing palladium-interlayer thickness up to a critical value and is always positive ($d < d_c$) or equal to zero ($d > d_c$). Therefore, the experimental results of our study indicate that there is an anisotropic exchange interaction in Co/Pd/CoNi multilayer films; these results can be described by introducing an anisotropic quasi-Heisenberg Hamiltonian

$$H = J_{12}^z(d_y) M_{1z} M_{2z} + J_{12}^\perp(d_y)(M_{1x} M_{2x} + M_{1y} M_{2y}). \tag{12}$$

Here, the FMR method is used to determine the value and form of the function $J_{12}^\perp(d_y)$ and the SWR method is used to determine the value and form of the function $J_{12}^z(d_y)$; the J components (as seen from the curves in Figs. 4, 5) may be characterized by completely different functional dependences.

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