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Ferromagnetic Resonance in a System of Magnetic Films with Different Curie Temperatures

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Abstract—The peculiarities of absorption of rf electromagnetic radiation (ferromagnetic resonance) in multilayer NiFe/$Ni_{0.65}Cu_{0.35}$(d)/CoFe structures in a wide temperature range are analyzed. It is shown that the type of interaction of the NiFe and CoFe ferromagnetic films via a "weak" ferromagnetic $Ni_{0.65}Cu_{0.35}$ interlayer changes from antiferromagnetic to ferromagnetic upon cooling and a decrease in interlayer thickness d. The detected temperature dependence of the interlayer interaction indicates the possibility of observation of a strong magnetocaloric effect in the structures under investigation.

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1. INTRODUCTION

The contact between two ferromagnets with different Curie temperatures leads to a substantial rearrangement of the magnetization distribution in the contacting subsystems. In the general case, this rearrangement involves demagnetization of the "strong" ferromagnet with a higher Curie temperature and magnetization of the "weaker" ferromagnet with a lower Curie temperature. An analogous situation takes place if one of the contacting magnets is in the paramagnetic phase. This "magnetic proximity" effect was investigated experimentally and theoretically in a number of studies [1–4]. The systems consisting of two strong ferromagnets (F) separated by an interlayer of a weak ferromagnet (f) with a Curie temperature lower or higher than the sample temperature are of considerable interest. As the material for such an interlayer, solid solutions Ni_xCu_{1-x} can be used, the Curie temperature of which can be controlled by varying concentration x of the solution [5, 6].

The interest in multilayer F/f/F structures is due to the possibility of designing microgenerators of microwave radiation on their basis [7], in which the mutual orientation of magnetic moments of ferromagnetic films substantially depends on temperature. High values of the efficiency of magnetic cooling for such systems were predicted theoretically in [8]. The exchange interaction at the interfaces between a paramagnetic films and ferromagnetic "banks" leads to a decrease in the magnetic moment of the paramagnetic film and ferromagnets in the case of antiparallel orientation of their magnetic moments and, hence, to an increase in the entropy of the system as compared to that for the parallel orientation. The switching between the states with different entropies occurs owing to the switching of the mutual orientation of magnetic moments of ferromagnetic films. In fact, the distribution of the "exchange" fields acting on the paramagnetic film changes. Since these fields are significantly stronger than the external magnetic field, the change in the entropy of the system is also large according to estimates. By analogy with the giant magnetoresistance effect, this effect can be referred to as the giant magnetocaloric effect.

The magnetic properties of F/f/F structures are determined by the effective interaction of ferromagnetic banks via the weak ferromagnetic interlayer. In turn, this interaction depends on the relation between the characteristic magnetic lengths and thicknesses of the films, temperature, and external magnetic field. The features of the magnetic moment dynamics in F/f/F structures (including the temperature dependences of the position and width of the ferromagnetic resonance (FMR) line) were studied theoretically and experimentally in [9, 10]. According to the reported results, the interlayer interaction is of the ferromagnetic type. In the present study, we will prove that such a behavior of the system is typical of only relatively small thicknesses of the weak ferromagnet. For large thicknesses of the interlayer and at high temperatures,

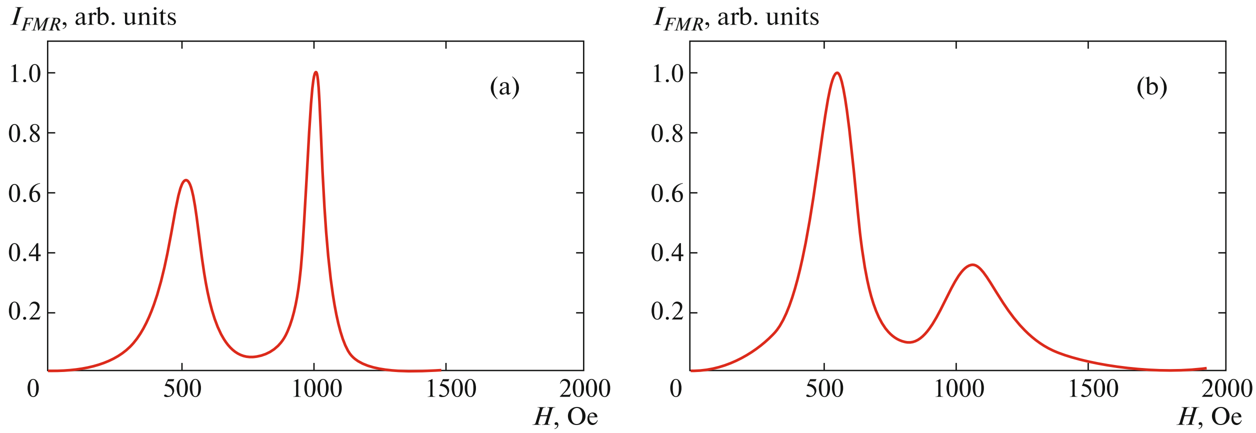

Fig. 1. (Color online) Dependences of microwave radiation absorption on the external magnetic field, recorded at room temperature for a structure with a weak ferromagnetic interlayer of thickness (a) $d = 7$ nm and (b) $d = 21$ nm.

the interlayer interaction is antiferromagnetic. Therefore, there exist a critical interlayer thickness and temperature at which the sign of the interlayer interaction in the F/f/F structures changes.

2. EXPERIMENTAL RESULTS

The multilayer $Ni_{80}Fe_{20}$(11 nm)/$Ni_{65}Cu_{35}$(d nm)/$Co_{60}Fe_{40}$(8 nm) structures with different thicknesses of the weak ferromagnetic interlayer (d = 7, 14, 21 nm) were obtained by high-vacuum magnetron sputtering (AJA 2200 multichamber system). The residual gas pressure did not exceed 10^{-7} Torr. The deposition was performed onto silicon and glass substrates. The structural and magnetic properties of the samples grown on different substrates did not differ qualitatively. The layer thicknesses were monitored by X-ray reflectometry (Bruker D8 Discover). According to our results obtained using transmission electron microscopy, the size of the crystallites in the investigated samples was 10–20 nm [11]. The films of $Ni_{65}Cu_{35}$ solid solutions were obtained by simultaneous sputtering of Ni and Cu targets, and their composition was determined by X-ray microanalysis using a dispersion spectrometer (INCA Energy Oxford Instruments). Static magnetic properties of the prepared films were determined by measuring the magnetooptical Kerr effect; the dynamic properties were studied using an EPR spectrometer (Bruker EMX 10/12) in the temperature range of 77–300 K; the working frequency of the spectrometer was 9.8 GHz, and the accessible range of a dc magnetic field was 0–15000 Oe. In all measurements, the dc and ac magnetic fields were oriented in the plane of the sample.

Figure 1 shows the dependences of the microwave absorption on the external magnetic field at room temperature for structures with $Ni_{65}Cu_{35}$ interlayer thicknesses d = 7 and 21 nm. The positions of the observed resonances are close to resonance fields of isolated NiFe films ($H_{res} \approx 1000$ Oe) and CuFe films ($H_{res} \approx 550$ Oe). However, the intensities of the peaks substantially depend on the thickness of the weak ferromagnetic interlayer. For the thin (7 nm) film (Fig. 1a), the intensity of the peak corresponding to the higher field (high-field peak) is higher, while for the interlayer with thickness of 21 nm (Fig. 1b), the peak corresponding to the lower value of the field (low-field peak) has a higher intensity. Obviously, such a behavior cannot be explained by the properties of isolated ferromagnetic films, and the observed resonances correspond to collective oscillations of magnetization in the multilayer system under investigation.

Figure 2 shows the temperature dependences of resonance fields for multilayer structures with a thin and a thick weak ferromagnet interlayer. It can be observed, first, that the resonance fields strongly depend on the temperature, which distinguishes the behavior of a multilayer system from the temperature dependences of resonance fields for isolated films. For example, the relative change in the position of the low-field resonance for the multilayer structure with the thin interlayer is 80% (Fig. 2b), while the change in the resonance fields for isolated CoFe and NiFe films upon a change in temperature from 130 to 300 K does not exceed 3%. Second, the temperature dependences of resonance fields for the structure with the thick interlayer are essentially nonmonotonic (Figs. 2c and 2d). The strongest change in the resonance fields occurs in the temperature interval of 240–270 K. It should be noted that the temperature dependences of resonance fields of the multilayer system with interlayer thickness d = 14 nm coincide qualitatively with the temperature dependences for the structure with the thin interlayer (d = 7 nm); for this reason, the graphs for this intermediate structure are not given here.

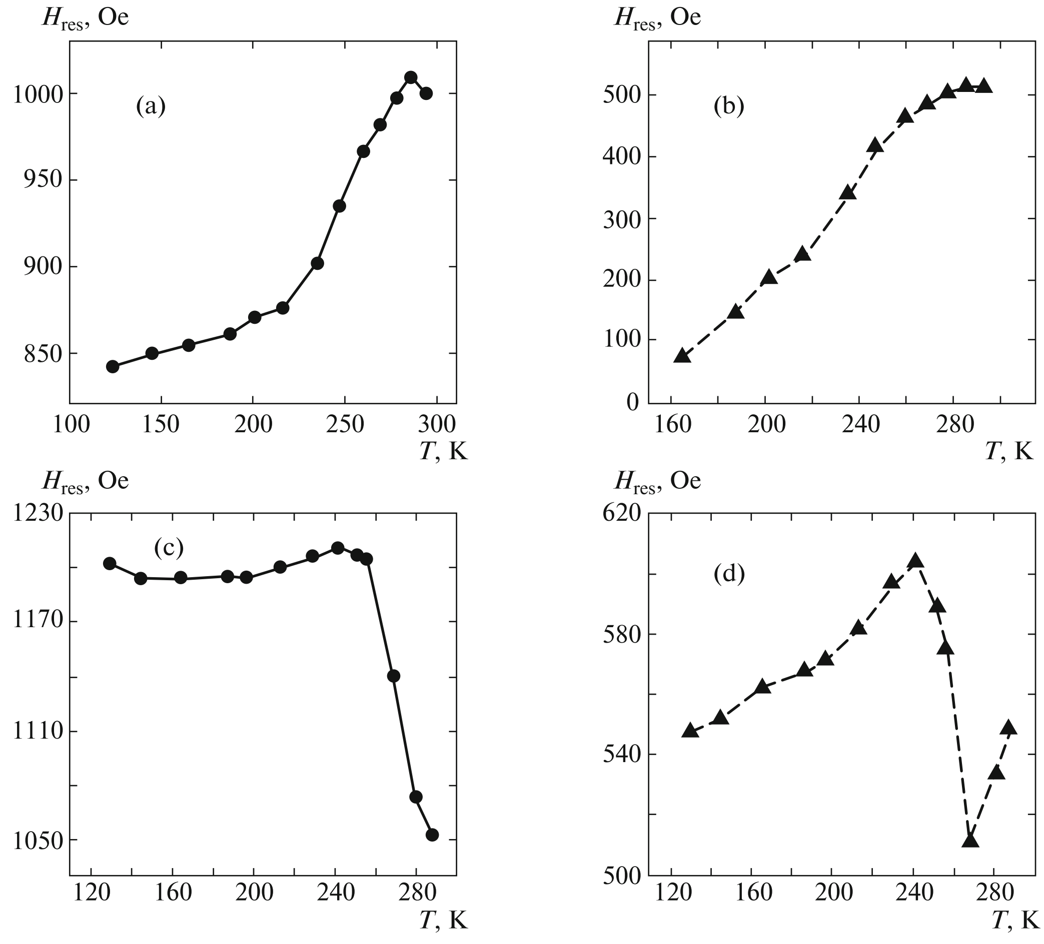

Fig. 2. Temperature dependences of resonance fields for (a, c) high-field and (b, d) low-field peaks for a multilayer structure with interlayer thickness d = 7 nm (a, b) and 21 nm (c, d).

Figure 3 shows the temperature dependences of the resonance peak amplitudes. The intensity of the high-field peak for the system with the thin interlayer is higher than the intensity of the low-field peak in the entire investigated temperature range. In the system with the thick interlayer, the relation between the intensities of the high- and low-field peaks changes at a temperature of about 250 K (Fig. 3b). Therefore, the main features of the FMR in the system under investigation lie in the sign reversal of the difference between the intensities of the high- and low-field peaks upon a change in the thickness of the weakly ferromagnetic interlayer and in temperature. The difference between these intensities changes sign for a certain critical thickness d_c of the interlayer, which lies in the interval 14 nm < d_c < 21 nm for the samples under investigation. For a system with $d > d_c$, the difference between the intensities changes its sign at critical temperature $T_C \approx 250$ K. It should be noted that these peculiarities were not detected before, which was probably due to the insufficiently wide range of thicknesses of the weak ferromagnetic interlayer in the systems studied in [10].

3. MODEL OF INTERLAYER INTERACTION AND DISCUSSION OF RESULTS

Let us assume that the main contribution to resonant absorption of microwave radiation of F/f/F structures comes from the oscillations of magnetic moments of the ferromagnetic banks (F). Then the two resonances in Fig. 1 correspond to synphase (acoustic branch) and antiphase (optical branch) oscillations of magnetic moments of ferromagnetic films. Clearly, the resonance intensity proportional to the absorbed power for the optical branch is lower than for the acoustic branch. In addition, the resonance field of the acoustic oscillation is higher ("ferromagnetic" interaction) or lower ("antiferromagnetic" interaction) than the resonance field of the optical oscillation depending on the sign of the interlayer interaction [12]. It follows hence that, for the critical thickness of the interlayer and at room temperature, the interlayer interaction reverses its sign; the ferromagnetic interaction corresponds to the small thickness of the interlayer and the antiferromagnetic interaction corresponds to the large thickness (see Fig. 1).

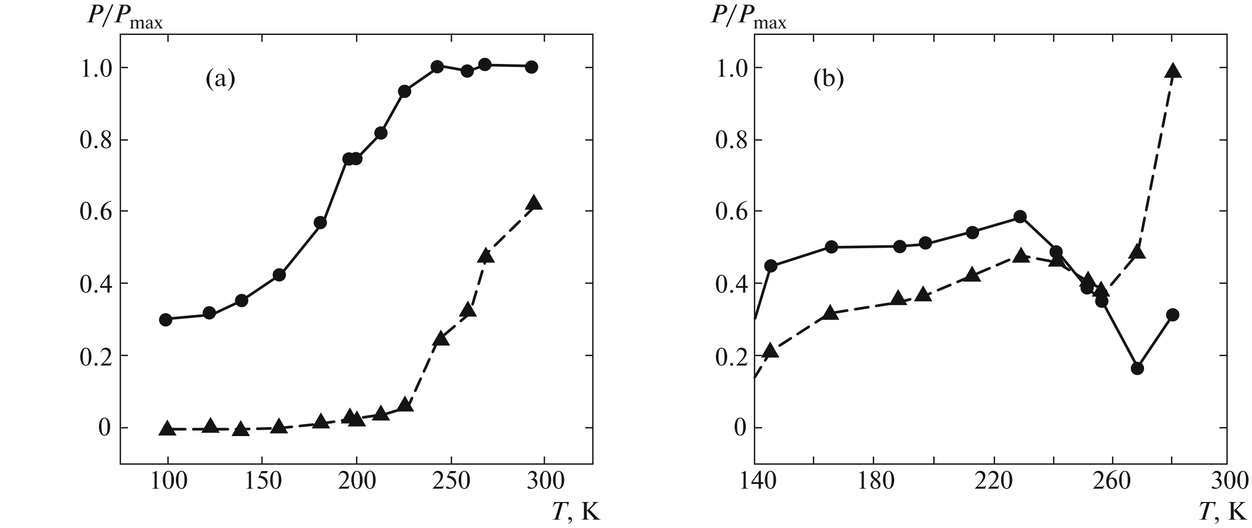

Fig. 3. Temperature dependences of the normalized amplitude of high-field (solid curves) and low-field (dashed curves) resonance peaks for a multilayer structure with interlayer thicknesses $d = 7$ nm (a) and 21 nm (b).

For the structure with $d = 21$ nm $> d_c$, the interlayer interaction reverses its sign upon a change in temperature, $T_C \approx 250$ K (see Fig. 3).

For describing the interlayer interaction in the F/f/F system, we write the free energy per unit volume in the form

$$F = \int\limits_{|z|<d/2}\left[\frac{1}{2}l_f^2\left(\frac{d\mathbf{m}_f}{\partial z}\right)^2 + \frac{1}{2}\tau\mathbf{m}_f^2 + \frac{1}{4M_f^2}\mathbf{m}_f^4 - \mathbf{H}\cdot\mathbf{m}_f\right]dz + \int\limits_{|z|>d/2}\left[\frac{1}{2}l_F^2\left(\frac{d\mathbf{m}_f}{\partial z}\right)^2 - \mathbf{H}\cdot\mathbf{m}_f\right]dz + \frac{1}{2}l_J(\mathbf{m}_F - \mathbf{m}_f)^2\Big|_{z=-d/2} + \frac{1}{2}l_J(\mathbf{m}_F - \mathbf{m}_f)^2\Big|_{z=d/2}, \tag{1}$$

where the z axis is perpendicular to the boundaries of the films, region $-d/2 < z < d/2$ is occupied by the weak ferromagnetic film f, and strong ferromagnet F lies outside this region. External magnetic field $\mathbf{H} = (H, 0, 0)$ is oriented along the x axis. We will further assume that the magnetic moments of the films lie in the xy plane, depend only on coordinate z, and do not produce demagnetizing fields. The first term in expression (1) is the free energy of the weak ferromagnet, which is written in the Landau form; $\tau = (T - T_C)/T_C$, T_C being the Curie temperature; and $l_f^2 \approx kT_C/aM_f^2$, a being atomic spacing [13, 14]. In this region, the magnetic moment $\mathbf{m}_f$ can change in magnitude as well as in direction. In the range of strong ferromagnets (second term in expression (1)), the magnetic moment changes only in direction, $\mathbf{m}_F = \mathbf{M}_F(\cos\theta, \sin\theta, 0)$, where θ is the angle between the magnetic moment and the external field and $\mathbf{M}_F$ is the saturation magnetic moment. The last two terms in expression (1) describe the exchange interaction of the weak antiferromagnetic film with the banks, which is characterized by constant $l_J > 0$ having the dimensions of length. The equations corresponding to the extremum of functional (1) and the boundary conditions for the magnetization of the weak ferromagnetic film have the form

$$\begin{gathered} l_f^2\frac{\partial^2\mathbf{m}_f}{\partial z^2} - \tau\mathbf{m}_f - \frac{1}{M_f^2}\mathbf{m}_f^3 + \mathbf{H} = 0, \\ l_f^2\frac{\partial\mathbf{m}_f}{\partial z} = \pm l_J(\mathbf{m}_F - \mathbf{m}_f)|_{z=\pm d/2}. \end{gathered} \tag{2}$$

For the magnetic moment distribution in strong ferromagnets, we obtain

$$\begin{gathered} \frac{\partial^2\theta}{\partial z^2} - \frac{1}{l_H^2}\sin\theta = 0, \quad l_H^2 = l_F^2\frac{M_F}{H}, \\ l_F^2\mathbf{M}_F^2\frac{\partial\theta}{\partial z} = \pm l_J\frac{\partial}{\partial\theta}(m_f^2 - 2\mathbf{m}_F\cdot\mathbf{m}_f)|_{z=\pm d/2}. \end{gathered} \tag{3}$$

The solution to Eq. (3) for "semi-infinite" ($d_F \gg l_H$, d_F is the thickness of the ferromagnet) ferromagnetic banks has the form

$$\cos\frac{\theta}{2} = \pm\tanh\frac{z \mp z_0}{l_H}, \tag{4}$$

where the upper and lower signs correspond to the right and left half-space, respectively, and constant z_0 can be determined from the boundary conditions.

To determine the character of the interlayer interaction, let us analyze the dependence of free energy (1) on the angle of rotation of the magnetic moments in the ferromagnetic banks. We assume that the magnetic moment at the boundaries of the weak ferromagnetic interlayer is codirectional with the magnetic moment of the ferromagnetic banks, which is characterized by angle $\theta(\pm d/2) = \pm\theta_0$. Assuming that the interaction at the boundaries is weak and the condition $l_J d \ll l_f^2$ is satisfied, we can write the solution to Eqs. (2) in the form

$$m_x = A + \frac{Bz^2}{2}, \quad m_y = Cz, \tag{5}$$

where the constants can be determined from the equation

$$A\left(\tau + \frac{2l_J}{d}\right) + \frac{1}{M_f^2}A^3 = H + \frac{2l_J M_F \cos\theta_0}{d} \tag{6}$$

and are defined by the formulas

$$B = \frac{2l_J(M_F\cos\theta_0 - A)}{dl_f^2}, \quad C = \frac{l_J M_F \sin\theta_0}{l_f^2}.$$

Substituting solutions (4)–(6) into Eq. (1) and assuming that angle θ_0 is small, we obtain the following expression for the free energy of the system:

$$F = M_F^2\theta_0^2\left(\frac{l_J^2\left\{l_f^2\left(\tau + \frac{2l_J}{d}\right) - \frac{d^2}{4}\left[\left(\tau + \frac{2l_J}{d}\right)^2 - \frac{\tau^2}{3}\right]\right\}}{\frac{d}{2}l_f^2\left(\tau + \frac{2l_J}{d}\right)^2} + l_F\sqrt{\frac{H}{M_F}}\right). \tag{7}$$

This expression was derived under additional conditions of smallness of the interaction at the film boundaries, $l_J \ll d$, and of the external magnetic field $H \ll M_F$, which allows us to disregard the terms linear in the magnetic field and to substantially simplify the expression for the free energy. In addition, we assume that the interlayer is in the paramagnetic phase ($\tau > 0$), and Eq. (6) has only the "induced" solution

$$A \approx \frac{l_J M_F \cos\theta_0}{d\tau/2 + l_J}.$$

Expression (7) implies that there exists a critical thickness d of the interlayer for which the parallel orientation of the magnetic moments of the ferromagnetic banks loses stability. This critical thickness can be determined from the condition of vanishing of the expression in the braces in Eq. (7). For $H = 0$, the critical thickness satisfies the equation

$$d_{c0} = 2l_f\sqrt{\frac{2l_J/d_{c0} + \tau}{(2l_J/d_{c0} + \tau)^2 - \tau^2/3}} \tag{8}$$

and has the following form in the limiting cases:

$$d_{c0} \approx \begin{cases} \frac{l_f^2}{l_J}, & l_J > \sqrt{\tau}l_f, \\ \sqrt{\frac{3}{2\tau}}l_f, & l_J < \sqrt{\tau}l_f. \end{cases} \tag{9}$$

Upon the application of a magnetic field, the critical thickness increases in accordance with the root law, $d_c \approx d_{c0} + \alpha\sqrt{H/M_F}$, where constant α can be determined from expressions (7) and (8).

The sign reversal of the interlayer interaction in an F/f/F system can be explained using the following considerations. If the interlayer is at a temperature higher than the Curie temperature, the free energy minimum corresponds to the demagnetized state (term $\tau\mathbf{m}_f^2$ in Eq. (1)). The tendency to a decrease in the mean square of the magnetic moment leads to "antiferromagnetic" ordering of the magnetic moments of the ferromagnetic banks. Indeed, the weak ferromagnetic film in this case is in zero (on the average) "exchange" field, and its magnetization decreases significantly. On the other hand, the "surface" terms responsible for the "proximity" effect lead to parallel orientation of the magnetic moments of the banks, for which the magnetic moment of the interlayer increases. The competition of the "bulk" and "surface" factors leads to the existence of the critical thickness of the interlayer. An increase in this critical thickness upon an increase in the external magnetic field is a consequence of the increase in the energy of ferromagnetic banks because of the formation of nonuniform distributions (4). When the temperature becomes lower than the Curie temperature of the interlayer, there are no factors leading to antiparallel orientation of the magnetic moments of the banks in the system. For this reason, the interlayer interaction at low temperatures ($T < T_C$, $\tau < 0$) must be "ferromagnetic" by nature for any interlayer thickness. Therefore, the model considered here explains qualitatively the main experimentally observed peculiarities of the FMR.

4. CONCLUSIONS

We have analyzed the peculiarities of the FMR in multilayer structures $NiFe/Ni_{0.65}Cu_{0.35}(d)/CoFe$ consisting of films with different Curie temperatures. It is shown that, at a temperature lower than the Curie temperature of the weak ferromagnet $Ni_{0.65}Cu_{0.35}$ ($T_C \approx 250$ K), the interlayer interaction depends on the interlayer thickness d; for a thick interlayer with $d > d_c$, the interaction is "antiferromagnetic," while for a thin interlayer ($d < d_c$), it becomes "ferromagnetic." The critical thickness of the weak ferromagnet for the investigated samples lies in the range 14 nm < d_c < 21 nm. The model considered here explains qualitatively the main features of the FMR observed in experiments. Our results confirm the substantial temperature dependence of the interlayer interaction and make it possible to determine the optimal parameters of the F/f/F system for the observation of the magnetocaloric effect.

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