ISSN 1063-7834, Physics of the Solid State, 2016, Vol. 58, No. 5, pp. 1034–1040. © Pleiades Publishing, Ltd., 2016.
Original Russian Text © G.S. Patrin, A.V. Kobyakov, I.A. Turpanov, K.G. Patrin, M. Rautskii, 2016, published in Fizika Tverdogo Tela, 2016, Vol. 58, No. 5, pp. 1000–1006.

SURFACE PHYSICS AND THIN FILMS

Magnetoresonance Properties of Three-Layer Co/Ge/Co Films

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Received May 27, 2015

Abstract—Three-layer Co/Ge/Co films have been studied using electron magnetic resonance. It has been established that the resonance spectrum of the film is a superposition of two Lorentzian lines. It has been found that the anisotropy induced at the cobalt–germanium interface makes the main contribution to the resonance spectrum and determines its features. The temperature dependences of the anisotropy field and the parameters of the interlayer exchange have been measured. The interlayer interactions exhibit an antiferromagnetic character and have been explained in terms of a model similar to the description of superexchange in magnetic dielectrics.

DOI: 10.1134/S1063783416050206

1. INTRODUCTION

Film structures in the ferromagnetic metal/semiconductor system attract attention, on the one hand, due to their possible applications and, on the other hand, due to the great variety of their physical properties [1, 2] As is known [3], the properties of film systems are substantially dependent on features of the technology of their preparation. For example, multilayer (Fe/Si) films demonstrated either ferromagnetic or antiferromagnetic interaction in the dependence on the substrate temperature [4]. The thermally induced behavior of the magnetization in $(Co/Si)_n$ films was studied in [5]. As the film that was initially cooled in a zero magnetic field (ZFC regime) was heated in weak magnetic fields at a certain temperature (blocking temperature T^*) that is dependent on magnetic field strength, the magnetic moment increases sharply. Similar behavior was observed in three-layer Co/Ge/Co films [6]. In this case, it was found that, in the dependence on film deposition rate and the substrate temperature at small thicknesses of the magnetic layer ($t_{Co} \leqslant 10$ nm), the system contained two magnetic phases, namely: the cubic cobalt matrix (fcc phase) contained granules of hexagonal cobalt (hcp phase) with mean sizes $r_{Co} \leqslant 2$ nm. The two phases precisely determine the temperature behavior of the magnetization [7]. The thermomagnetic properties of the films were explained in terms of the generalized Stoner–Wolhfarth model [8], in which the hexagonal granules are represented as quasi-Izing particles that are randomly distributed in a magnetic isotropic cubic matrix and bounded to it by exchange interaction. In a number of cases, this behavior is of a practical interest, since it makes it possible to control the properties of the whole film structure at the technological stage and also by external actions.

However, it is impossible to obtain detailed real information on the interlayer interactions in such multilayer films using only quasi-static measurements, because the existence of the two phases shields all details, particularly at temperatures $T < T^*$. The electron magnetic resonance (EPR) method makes it possible to solve this problem. In processes of the magnetic dynamics, each magnetic subsystem has a proper vibration frequency that is sensitive to changes in the internal magnetic fields [9]. Because of this, we used EPR method to determine the interlayer interactions in three-layer Co/Ge/Co films.

2. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUE

We synthesized Co/Ge/Co films with various averaged-in-area thicknesses of the nonmagnetic germanium layer. The films were deposited by ion–plasma sputtering at a basis pressure of $P = 10^{-6}$–10^{-7} Torr in an argon atmosphere [7]. The substrate material was a glass; during deposition, the substrate temperature was $T \approx 373$ K. We studied a series of the films with $t_{Co} = 13 \pm 0.3$ nm. The germanium thickness was variable; the cobalt deposition rate was 0.15 nm/s, and that of germanium was about 0.12 ± 0.02 nm/s. At these technological conditions, the cobalt exists preferably in the metastable cubic phase, and it exists in the hexagonal phase if the rate is an order lower. The averaged thicknesses of germanium and cobalt layer were found using X-ray spectroscopy.

The magnetic dependences were measured using an MPMS XL SQUID magnetometer in fields to 50 kOe. The electron magnetoresonance spectra were measured using a "Bruker E 500 CW ERP" spectrometer operating at the frequency ω_{MWF} = 9.2 GHz. In the experiment, the microwave and dc bias magnetic fields laid in the film plane. The measurements were performed in the temperature range 100–500 K.

3. EXPERIMENTAL RESULTS AND DISCUSSION

Before analyzing the results of the magnetoresonance measurements, we note that the magnetization of a pure cobalt film in the fcc phase, as can be seen from Fig. 1, depends strongly on the film thickness in the range of small thicknesses ($t_{Co} \leqslant 6$ nm) and reaches saturation at thicknesses $t_{Co} \geqslant 10$ nm. The film had no magnetic moment, as the cobalt layer thickness was less than 0.5 nm. These data were obtained on the polycrystalline films in the magnetic field $H = 30$ kOe lying in the film plane. A similar situation was observed in the multilayer structures of the ferromagnetic metal/semiconductor system. For example, the magnetic moment disappeared in films $(Co/Si)_n$ at a thickness of $t_{Co} \sim 1.2$ nm [10] and in the $(Co/Ge)_n$ films at a thickness of $t_{Co} \sim 2.0$ nm, which seems to be due to the formation of a "dead layer." The cobalt thickness of 13 nm was chosen from the following considerations: first, in order that the cobalt layer magnetization was stable during uncontrolled changes in the magnetic layer thickness, and, second, in order that the Zeeman interaction was as weak as possible and did not shield the interlayer interaction.

The electron magnetic resonance spectrum of Co/Ge/Co films with a nonmagnetic interlayer had an unusual shape. The studies were carried out at temperatures $T > T^*$ when a nonzero magnetization appeared in the films. Figure 2 shows the typical magnetic resonance spectra of the film with $t_{Ge} = 9$ nm. As is seen, the initial spectrum was an individual absorption line, but the spectrum became complex at higher temperatures. The observed spectrum was approximated well by a superposition of two Lorentzian-type lines. Figure 3 shows the temperature dependences of the values of the magnetic fields. It is seen that the low-field resonance (line *1* in Fig. 2) appeared only at a temperature higher than T^*, while the high-field resonance (line *2* in Fig. 2) was observed over the entire temperature range under study. As follows from an analysis of the magnetization (Fig. 3 in [8]), the increase in the fraction of the hexagonal cobalt fraction led to the increase in temperature T^* at which the magnetization appeared in low fields. The increase in the magnetic field significantly deceases at T^*. The scheme of analyzing the behavior of the resonance

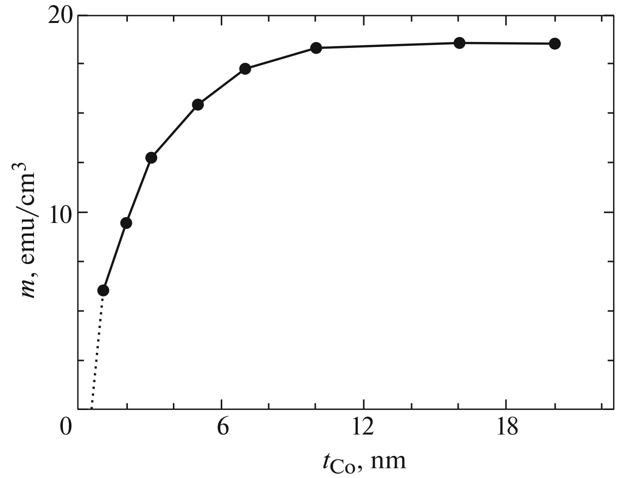

Fig. 1. Magnetization of cobalt vs. film thickness at T = 300 K.

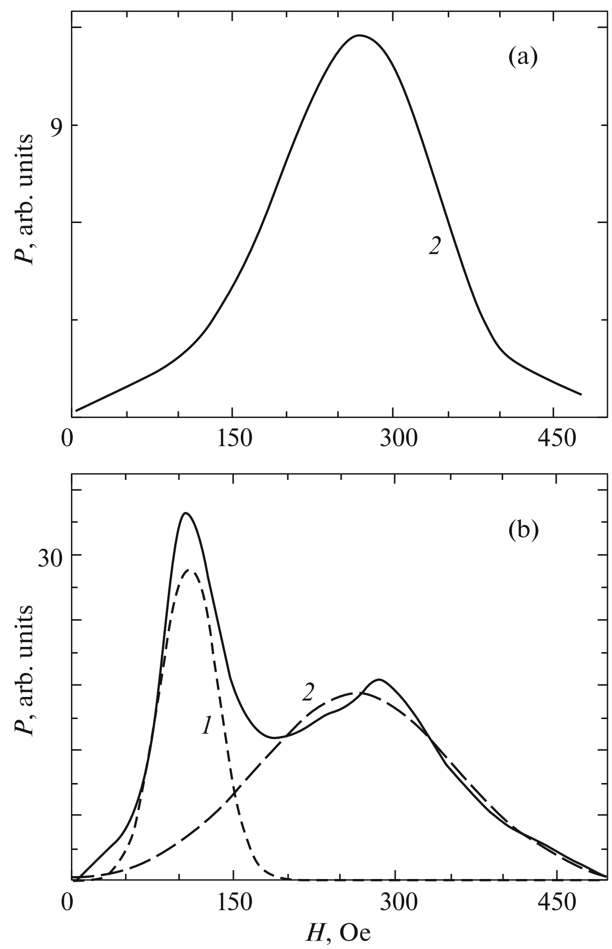

Fig. 2. Magnetic resonance spectra for the film with t_{Ge} = 9 nm measured at T = (a) 120 and (b) 340 K. Curves *1* and *2* are the Lorentzian-type lines.

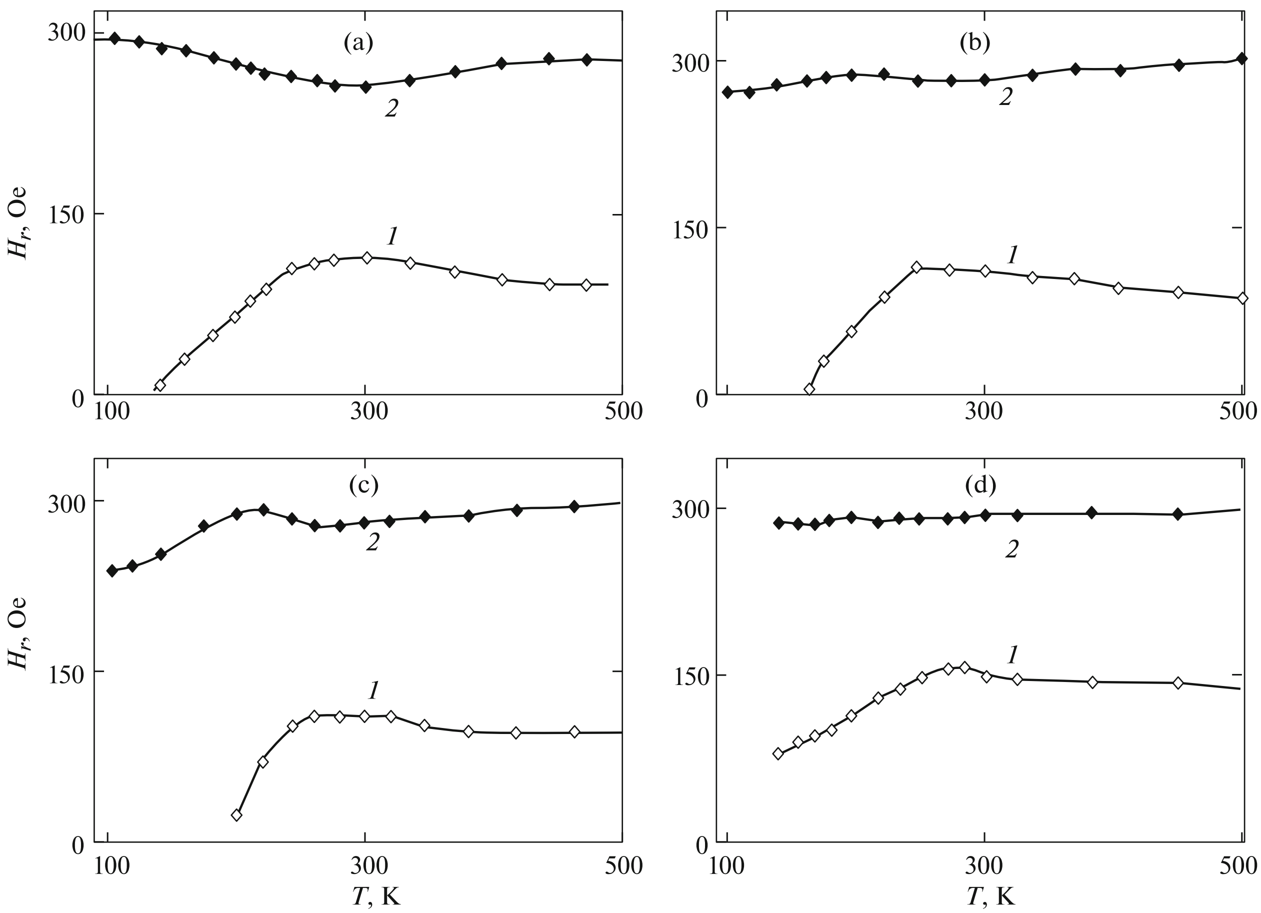

Fig. 3. Temperature dependences of the resonance fields for the films with thicknesses t_{Ge} = (a) 6, (b) 9, (c) 15, and (d) 18 nm.

parameters in the films studied was based on the comparison of the values of the resonance fields and thermomagnetical behavior of the magnetization. The temperature behavior of the magnetization of two-phase Co/Ge/Co films was studied in detail in [7, 8]. Thus, we can suggest that the magnetization of each of the magnetic layer was determined. Note that the magnetization almost attainment to saturation in fields ~300 Oe over the entire temperature range. In this case, no anisotropy of the resonance field in the film plane was observed.

First, we should define the magnetic structure of the three-layer system. The following variants are possible.

(i) If the ferromagnetic layers are not bounded by the exchange interaction, then, because of the identity of the layers, this must lead to the existence of either an individual line or two close lines of microwave absorptions with close parameters and similar behavior with varying temperature. However, this situation was not observed in the experiments.

(ii) As the ferromagnetic interlayer interaction exists, two resonance peaks are possible [9]. One of them is related to the uniform resonance and is determined by the Kittel formula with the inclusion of the magnetic crystallographic anisotropy, and another is due to the resonance in the state with a domain structure. Since the ferromagnetic layers were polycrystalline, the width of the resonance peak corresponding to a nonuniform state was as large as that which could not be observed in the condition of our experiments (below, we present the anisotropy parameters).

(iii) As a working variant, we use a model of antiferromagnetic bond between the layers.

To justify our approach to the analysis of the experiments on the measurements of the magnetic resonance and to illustrate the correlation of the magnetic characteristics and the resonance properties, we present typical data obtained on the film with $t_{Ge} = 9$ nm. Figure 4 shows the field dependence of the magnetization measured in magnetic fields $|H| \leqslant 40$ kOe for the film with $t_{Ge} = 9$ nm at $T = 150$ K. The magnetization increases linearly and insignificantly (~4% of the value in the saturation field) over the entire range of the magnetic fields. This testifies that the "paraprocess" of magnetization still takes place because of the existence microregions, centers of which can be chaotically oriented granules of strongly anisotropic hexagonal cobalt. The insert shows the hysteresis loops

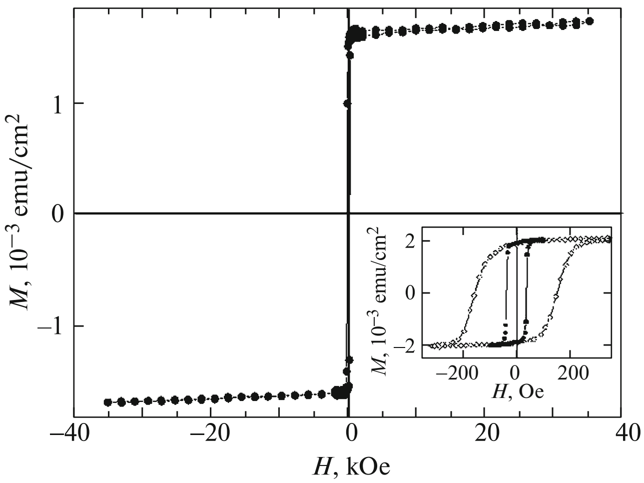

Fig. 4. Field dependences of the magnetization of the films with $t_{Ge} = 9$ nm at $T = 150$ K. The insert shows the open hysteresis films at temperatures $T = 150$ and 280 K (bright and dark symbols, respectively).

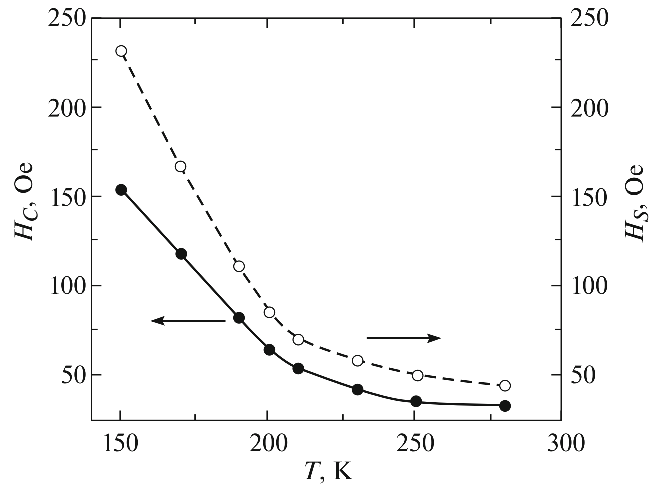

Fig. 5. Temperature dependences of coercive force H_C and saturation field H_S of the film with thickness $t_{Ge} = 9$nm.

measured at temperatures $T = 150$ and 280 K. It is seen that, as temperature increases, the loop became more "rectangular" along with decreasing coercive force H_C. Figure 5 shows the temperature dependences of coercive force H_C and saturation magnetization field H_S in the temperature range under study. It follows from Fig. 5 that the resonance fields corresponding to the dependences of the *1* type (Fig. 3) got in the saturation region at temperatures $T > 200$ K.

Since the features of the magnetic behavior were determined by strongly anisotropic granules of the hexagonal (impurity) phase, we describe their influence on the magnetoresonance properties using effective anisotropy K_A. In this case, the free energy per unit area of the Co/Ge/Co film with the antiferromagnetic order is [12]

$$U = -J\cos(\varphi_1 - \varphi_2) - t_{Co}\{\mathbf{H}(\mathbf{m}_1 + \mathbf{m}_2) + (K_A/(2m^2))[m_{Z1}^2 + m_{Z2}^2] + 2\pi[m_{Z1}^2 + m_{Z2}^2]\}, \quad (1)$$

where J is the constant of interlayer interaction, $\mathbf{H}$ is the external magnetic field (it lies in the film plane in is directed along axis X), $\mathbf{m}_i = \mathbf{M}_i/t_{Co}$ is the averaged magnetization, $\mathbf{M}_i$ is the magnetic moment of unit area of the ith ferromagnetic layer, φ_i is the magnetization angle in a plane counted from axis Y, index $i = 1$, 2 numbers the magnetic layers, $m_1 = m_2 = m$, K_A is the constant of the effective anisotropy, t_{Co} is the magnetic layer thickness, and axis Z is perpendicular to the film plane. We assumed in our calculations that the film is in an unsaturated state. In this case, assuming that both ferromagnetic layers are identical, at the condition of equilibrium in fields lower than the saturation field, at the antiferromagnetic interlayer interaction, we have $\varphi_1 = \pi - \varphi_2 = \varphi$, which gives

$$\sin\varphi = H/(2H_J), \quad (2)$$

where $H_J = J/(t_{Co}m)$.

Condition $H_S = 2H_J$ corresponds to the state when the layer magnetizations become parallel. At these conditions, the resonance frequencies of the layers are described by expressions [12]

$$(\omega_1/\gamma)^2 = H\{H + (H_A + H_M)[H/(2H_J)]\}, \quad (3)$$

$$(\omega_2/\gamma)^2 = [2H_J(H_A + H_M)]\{1 - [H/(2H_J)]^2\}, \quad (4)$$

where

$$H_A = K/(2m), \quad H_M = 4\pi m, \quad (5)$$

and γ is the giromagnetic ratio.

First and foremost, note that the expression for the acoustic mode frequency (3) converses to the Kittel formula at the saturation magnetization ($H/(2H_J) = 1$) and the optical mode frequency (4) becomes zero. Now, we should find the correlation between solutions (3) and (4) and lines *1* and *2* in Fig. 3. From the experiment, it follows (Fig. 4) that the magnetization of one layer is $m = 807$ Oe, and we obtain $H_M = 10.14$ kOe.

When fitting parameters H_J and H_A, we calculated both variants: (a) line *1* corresponds to the acoustic mode; (b) line *1* corresponds to the optical mode. In the case, when line *1* was ascribed to the acoustic mode and line *2* to the optical mode, we obtained that H_A must be ~500 kOe, which is quite far from the real scale of the values observed in the experiments. In the case when line *1* was identified as the optical mode and line *2* as the acoustic mode, we obtained H_A ~ 30–35 kOe that are at least comparable with H_M. Thus, this is the situation for which we calculated exchange parameters H_J and the value $H_\Delta = [2H_J(H_A + H_M)]^{1/2}$ that characterizes the optical branch in the magnetic resonance spectrum.

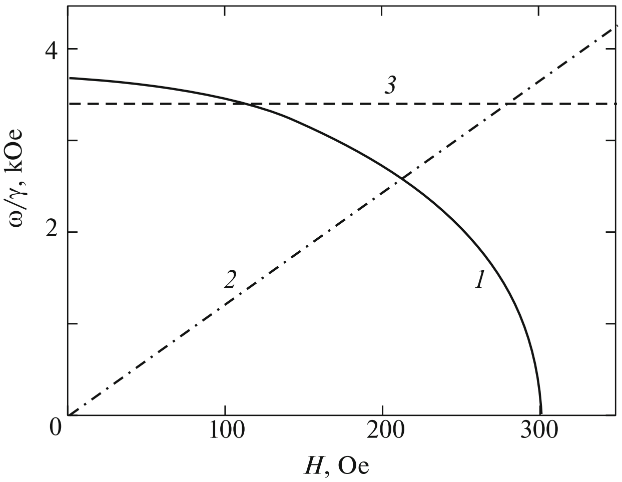

Fig. 6. Calculated frequency–field dependences of the magnetic resonance of the Co/Ge/Co film; t_{Co} = 13 nm, t_{Ge} = 9 nm, T = 300 K: (*1*) optical mode for the antiferromagnetic exchange (calculation by Eq. (4)), (*2*) acoustic vibration mode (calculation by Eq. (3)), and (*3*) the resonance frequency (in the field units).

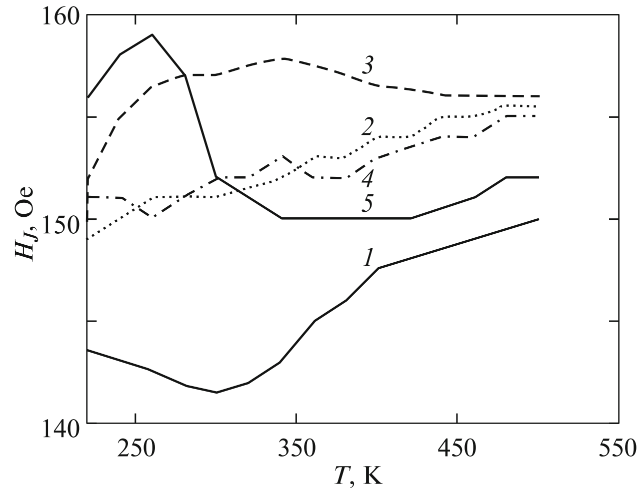

Fig. 7. Temperature dependences of the exchange field in the Co/Ge/Co films with antiferromagnetic interlayer interaction; t_{Ge} = (*1*) 6, (*2*) 9, (*3*) 12, (*4*) 15, and (*5*) 18 nm.

For illustration, we calculated the frequency–field dependences of the resonance for the film with the germanium thickness t_{Ge} = 9 nm at T = 300 K. Figure 6 shows the corresponding dependences. Here, line *3* corresponds to the experimental value of ω/γ, and line *2* is determined by dependence (3). As follows from the fitting using Eq. (3), the anisotropy field was H_A = 34.26 kOe. Since the magnetization of the films for magnetic fields, at which the resonance absorption took place, attained the saturation at temperatures $T \geqslant$ 200 K, all the parameters had the same values for both Eq. (3) and Eq. (4) in this temperature range. These were temperatures for which we performed the fitting of the experimental data in Fig. 3 by calculating H_J and H_A.

The results of the fitting H_J are shown in Fig. 7. Attention is drawn to the fact that, for all the films, the exchange field corresponded to the antiferromagnetic interlayer interaction and, in this case, the value of H_J slightly (~0.5%) oscillated at constant temperature, depending on the nonmagnetic layer thickness. At temperatures T > 400 K, the values of H_J differed slightly (less than by 0.2%) and seem to tend to a value close to the value for the film with t_{Ge} = 18 nm.

As is known [1], the oscillating dependence on the interlayer thickness is typical of the interlayer interaction in multilayer magnetic films with a nonmagnetic interlayer. The oscillation period is, as a rule, 1–3 nm; however, all film imperfections (surface roughness, chemical heterogeneity at the interface etc.) increase the oscillation period and their smoothing [13]. However, there are systems and mechanisms that demonstrate the nonoscillating type of the interlayer interaction [14, 15]. In terms of the model from [15], when magnetic layers are represented by narrow *d* bands that are shifted by the value $\Delta^{\uparrow,\downarrow}$ with respect to Fermi energy ε_F of all system, and nonmagnetic layers are represented by bands of free conduction electrons, two contributions into the interlayer exchange can be separated. One of them is the RKKY (Ruderman–Kittel–Kasui–Yosida) contribution describing the exchange interaction between magnetic ions via collectivized conduction electrons; another is the antiferromagnetic interlayer exchange of the "superexchange" type that takes place in magnetic dielectrics. At condition $k_F t_{NM}(\Delta^{\uparrow,\downarrow}/\varepsilon_{Fe}) < 1$ (here, k_F is the wave vector at the Fermi level and t_{NM} is the nonmagnetic layer thickness), which is possible if the value $k_F t_{NM}$ is small or $\Delta^{\uparrow,\downarrow}/\varepsilon_{Fe} \ll 1$, the antiferromagnetic exchange prevails and the RRRY contribution is negligibly small.

This approach makes it possible to explain our experimental results. Figure 8 shows a fragment of the energy band structure of the Co/Ge/Co film that illustrates the model situation. In the case of a semiconductor interlayer, the Fermi level is in the forbidden band, and then the existence of the conduction electrons in this band near the Fermi level can be due to the formation of the band "tails" in the semiconductor because of the existence of a disorder or the existence of magnetic impurities. At the thicknesses of the semiconductor in the Co/Ge/Co films used in this work, germanium was partially in an amorphous phase [6], which led to formation of the "tails." In this case, the electron density was sufficient to transfer the interaction between the ferromagnetic layers (a similar scenario was proposed for the description of the magnetic properties in Fe/Si/Fe films at low temperatures [16]).

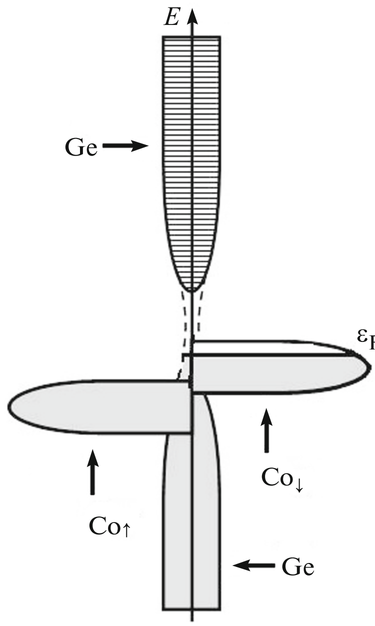

Fig. 8. Fragment of the energy band structure of conduction electrons in the Co/Ge/Co film. Co↓ and Co↑ denote the 3d bands with corresponding directions of spins. The dashed line show the band "tails" of the semiconductor.

The decrease in the conduction electron density in a nonmagnetic interlayer leads to significant weakening of the RKKY-type interaction between the magnetic layers. In this case, the relative fraction of the contribution of the antiferromagnetic interaction increases.

Figure 9 shows the calculated temperature dependences of H_Δ. We focus our attention on the existence of the maximum in the vicinity of temperature $T \approx$ 300 K. As well as for the value of H_J, there are oscillations in the dependence on the thickness of the semiconductor interlayer. It is clear that such behavior of H_Δ is mainly determined by the behavior of the magnetic anisotropy of the film structure. As is known [17], the magnetic anisotropy of the film structure consists of two parts: one of them is determined by the contribution of the volume anisotropy of a material K_V and another is the contribution of the surface anisotropy K_S (or anisotropy at interface); i.e., $K_A = K_V + K_S$. In thick films, K_V dominates, while, as the magnetic layer thickness decreases, the influence of anisotropy K_S at the surface or at the interface comes to dominate. As was noted in [17], the surface contribution can be an order higher than that of the volume anisotropy. In hexagonal cobalt, $K_V \sim 5 \times 10^6$ erg/cm^3 [18], which gives anisotropy field H_A of several kilooersted. This allows us to suggest that the main contribution to the anisotropy of a film structure is due to the cobalt–germanium interface and, as follows from our experiment, the surface anisotropy is the easy-plane anisotropy ($K_A > 0$).

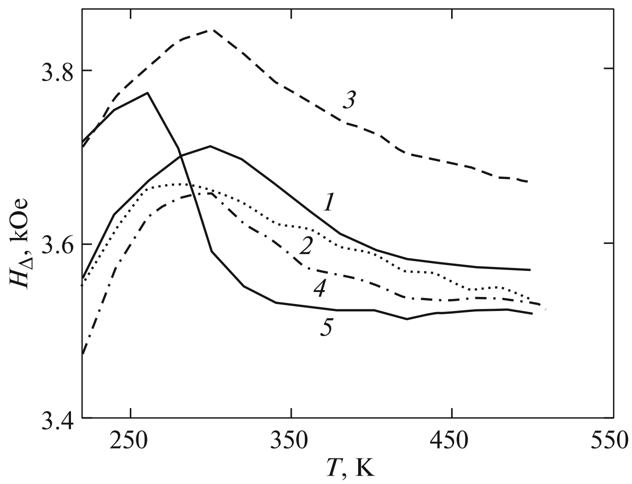

Fig. 9. Temperature dependences of the gap width in the resonance spectrum of the films with t_{Ge} = (*1*) 6, (*2*) 9, (*3*) 12, (*4*) 15, and (*5*) 18 nm.

Thus, it was found for the Co/Ge/Co structure that the existence of anisotropic granules of hexagonal cobalt determines the processes of magnetization of ferromagnetic cobalt layers and the interface anisotropy provides the fact that the magnetization lies in the film plane and determines the features of its resonance behavior.

4. CONCLUSIONS

As a result of the magnetoresonance studies of the Co/Ge/Co films, it was found that the interlayer interaction is long-range, and the values and the sign of the interlayer interaction were determined. For example, the nonmagnetic interlayer thicknesses at which the interlayer exchange is observed can be ~10 nm. These large thicknesses are not typical of the metallic or dielectric (tunneling) interlayers. On the other hand, in the case of semiconducting or semimetallic materials, the mean free path of electrons can be large, and it explains the nonmagnetic interlayer thicknesses at which the interlayer interaction is still observed (for example, in the CoFe/Bi/CoFe structure, the oscillation period was ⩾10 nm [19]).

It is clear that the abovementioned explanation is qualitative. Nevertheless, the mechanism described in [15] encloses the main features of the interlayer interactions. More detailed description of the exchange interactions and the magnetic anisotropy in the ferromagnetic metal/semiconductor system requires taking into account a real electronic structure and, what is not less important in the light of recent studies [11, 20], the interface structure, since the interface thickness is an nanometers, and new phases form in the interface. All that is mentioned above determines the directions of future studies.

ACKNOWLEDGMENTS

This study was performed in the framework of the State Task of the Ministry of Education and Science of the Russian Federation to the Siberian Federal University (task no. 3.2534.2014/K) and was supported by the Russian Federation for Basic Research (project no. 14-02-00238-a).

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Translated by Yu. Ryzhkov