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*In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.*
Magnetic and superconducting heterostructures in spintronics†

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This paper is a brief review of investigations, which were carried out during last years by team of magnetic nanostructures and spintronics laboratory, and is dedicated to the 80th anniversary of our Teacher - professor B.I. Kochelaev.

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1. Introduction

An electron is characterized by charge \(e\) and spin \(s = 1/2\). Spintronics is a new approach to electronics which use control of electron transport through the spin of electron in addition to its charge for device functionality (see, for example, [1–3]).

The basic elements of spintronics are magnetic, superconducting and tunneling heterostructures. In this paper we present results of studies, which were carried out during last years by the team of Physics of Magnetic Nanostructures and Spintronics laboratory (FMNS).

2. Model of point contact and basics of the theory

In the present work a model of the point contact (PC) between two ferromagnetic metals (see, for example, Refs. [4, 5]) with different conduction properties of the spin sub-bands is considered. The PC is simulated by a nanosize circular hole of the radius \(a\) made in an impenetrable membrane, which divides the space on two half-spaces occupied by single-domain ferromagnetic metals (see Fig. 1). The \(z\)-axis of the coordinate system is chosen to be perpendicular to the membrane plane. A model of linear domain wall has been used to account for the finite contact length. When magnetizations on both sides of the contact is in parallel (P) alignment, there is no domain wall in the constriction, and the electric current flows through the contact independently in each of the conduction electron spin-subbands. At antiparallel (AP) alignment of the magnetizations, a domain wall is created in the constriction [6]. Simultaneously, the conduction spin-subband assignment in one of the magnetic domains reverses with respect to the previous one. In the case of a ferromagnetic PC, the band structures of the spin-subbands of the ferromagnetic metals do not coincide for either spin-up or spin-down conduction electrons. It is obvious that the potential barriers at the interface of the contact are different for the P and AP alignments. As a result, scattering of electrons associated with these potential barriers and magnetization profiles at the interface are different for the two alignments, which gives rise to magnetoresistance.

The electron motion on the both sides of the contact can be described by transport equations for quasi-classical Green functions (GF’s) [7]. These GF’s are symmetrical and antisymmetrical with respect to \(z\) projection of the quasiparticle momentum and satisfy Boltzmann equations in

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Figure 1. Schematic view of short channel with domain wall is shown. The arrows denote parallel and antiparallel alignments of magnetization FM$L$ and FM$^R$ ferromagnetic.

The τ-approximation. We develop a theory of electric transport through magnetic PC’s taking into account gradient terms in the series expansion of GF’s. The theory covers ballistic $l > a$ and diffusive $l < a$ regimes ($l$ is the mean free path) to explain the variety of observed experimental data.

The case of the same ferromagnetic metal was considered in [7] in the quasi-classical approximation. Using the same approach, we give a general derivation of conductance for different ferromagnetic metals. Our aim is to calculate the spin-polarized current $I^z_\alpha$ through the hole in response on the voltage drop $V$ applied to the outer leads far away from the contact:

$$I^z_\alpha (z \to 0, t) = a \int_0^\infty dk J_1 (ka) j^z_\alpha (0, k, t),$$

(1)

where $\alpha = (\uparrow, \downarrow)$ is the spin index. Here the Bessel function $J_1 (ka)$ comes from the integration of the current density $j^z_\alpha (z = 0, \rho)$ over the circular contact cross-section, and $j^z_\alpha (0, k, t)$ is the Fourier transform of the current density $j^z_\alpha (z = 0, \rho)$ over the in-plane coordinate $\rho$. The current density can be expressed via the antisymmetric quasiclassical GF, $g_{a,\alpha} (0, k, t)$, as follows ($k_B = \hbar = 1$):

$$j^z_\alpha (0, k, t) = - \frac{e p_{F,\alpha,\text{min}}}{2\pi} \langle \cos \theta_{L,\alpha} g_{a,\alpha} (0, k, t) \rangle_L,$$

(2)

where $p_{F,\alpha,\text{min}}$ is the Fermi momentum, which is the smallest of the momentums ($p_{L,\alpha} F, p_{R,\alpha} F$), $\theta_{L,\alpha}$ is the angle between the $z$ axis and the direction of movement of the electron to the hole, $g_{a,\alpha} (0, k, t)$ is antisymmetrical with respect to $z$ projection of the quasiparticle momentum and satisfies Boltzmann equations in the $\tau$ approximation:

$$l^z_{z,\alpha} \frac{\partial g_{a,\alpha}}{\partial z} + (1 - i \mathbf{k} \cdot \mathbf{l}_{||,\alpha}) g_{s,\alpha} = \langle g_{s,\alpha} \rangle,$$

(3)

$$l^z_{z,\alpha} \frac{\partial g_{s,\alpha}}{\partial z} + (1 - i \mathbf{k} \cdot \mathbf{l}_{||,\alpha}) g_{a,\alpha} = 0.$$

(4)

Here $g_{s,\alpha}$ is the symmetric GF, $\mathbf{l}_{||,\alpha}$ is the vector defining the electron mean free path of conduction in the contact plane whose absolute value is determined by the geometry: $l^2_{||,\alpha} = l^2_{z,\alpha} - l^2_{\alpha}$, where $l_{z,\alpha} = l_{\alpha} \cos \theta_{\alpha}$ is the projection on the axis $z$, $\mathbf{k}$ is the wave vector in the contact plane. The angular brackets in (2) and (3) mean averaging over the solid angle:

$$\langle g_{s,\alpha} \rangle = \frac{1}{2\pi} \int_0^{\pi/2} \int_0^{2\pi} g_{s,\alpha} \sin \theta d\theta d\varphi.$$

GF’s in the Boltzmann equations (3) and (4) satisfy boundary conditions [8]:

$$g^L_{a,\alpha} (0) = g^R_{a,\alpha} (0) = \begin{cases} g_{a,\alpha} (0), & p_{||,\alpha} < p_{F,\alpha}^L, p_{F,\alpha}^R, \\ 0, & p_{||,\alpha} > \min \left[p_{F,\alpha}^L, p_{F,\alpha}^R\right]. \end{cases}$$

(5)
2R_\alpha g_{s,\alpha}(0) = D_\alpha (g^L_{s,\alpha}(0) - g^R_{s,\alpha}(0)), \quad (6)

where \(p_{\parallel,\alpha}\) is the projection of the spin-dependent Fermi momentum \(p_{F,\alpha}\) on the plane of the contact, \(D_\alpha\) and \(R_\alpha = 1 - D_\alpha\) are the angular- and spin-dependent quantum-mechanical transmission and reflection coefficients, respectively. Boundary conditions (5) and (6) obey the specular reflection law:

\[ p_{\parallel,\alpha} = p_{F,\alpha} \sin \theta_{L,\alpha} = p_{F,\alpha} \sin \theta_{R,\alpha}. \quad (7) \]

3. Spin-polarized current in ferromagnetic point contact

Solutions of equations (3) and (4) together with boundary conditions (5) and (6) lead to an expression for the spin-polarized current through PC. The basic mathematical background and calculation details can be found in articles [7,9]. We develop the theory [7,9] of electric transport through magnetic PC’s using the following series expansion of the GF:

\[ g^{L(R)}_{s,\alpha}(z, \varepsilon) \approx \tanh \left( \frac{\varepsilon}{2T} \right) + f^{L(R)}(0, \varepsilon) + z \cdot \frac{\partial f^{L(R)}(z, \varepsilon)}{\partial z}. \quad (8) \]

Here the first term corresponds to equilibrium GF \(g^{eq}_{s}\) far away from the contact, \(\varepsilon\) is the energy of electron, \(T\) is the temperature. The second term corresponds to the first order in the expansion of the GF and determines the current heterogeneity in the contact plane. The third term in the expansion for GF corresponds to gradients of the chemical potential. Solution of (3) and (4) taking into account all the terms in the expansion (8) leads to a rather cumbersome expression for the components to the spin current. Here we write it in the simplified form

\[ I_{\alpha}^{z} = \frac{e^2 n_{F,\alpha,\min} a^2 V}{2\pi} \int_{0}^{\infty} dk J_{1}^{2}(ka) \cdot F_{\alpha}(k, \theta_{L,\alpha}), \quad (9) \]

where \(F_{\alpha}(k, \theta_{L,\alpha})\) represents the sum of functional dependencies and the integrals of the transmission coefficients \(D_{\alpha}\) of the domain wall and parameters \(i^{L(R)}_{\alpha}\), \(p^{L(R)}_{F,\alpha}\) of the metals in the contact. We write \(F_{\alpha}(k, \theta_{L,\alpha})\) in a form of three terms

\[ F_{\alpha}(k, \theta_{L,\alpha}) = \langle \cos \theta_{L,\alpha} D_{\alpha} (\cos \theta_{L,\alpha}) \rangle_{L} + F_{\alpha}^{\text{heter}}(k, \theta_{L,\alpha}) + F_{\alpha}^{\text{grad}}(k, \theta_{L,\alpha}), \quad (10) \]

where the first term can be used to interpret the spin-polarized conduction in planar contacts. The second term in (10) can be used to calculate the conductivity at a nonuniform current distribution in the plane of the point contacts. The third term \(F_{\alpha}^{\text{grad}}(k, \theta_{L,\alpha})\) accounts for the chemical potentials bending at the borders of the heterostructure. Notice finally that the current given by equation (9) refers to a particular spin-channel of conductance. The total current through the nanocontact is the sum of currents for both spin-channels. The formal expression for the second channel is the same, but with all physical parameters referred to the second spin-channel.

4. Magnetoresistance of magnetic point contacts

The total current through a magnetic PC combines two spin-channels for P and AP mutual orientations of magnetizations in the magnetic domains. Then, magnetoresistance (MR) is characterized by a dimensionless ratio:

\[ \text{MR} = \frac{I^{P} - I^{AP}}{I^{AP}}, \quad (11) \]
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Figure 2. (Color online) Dependence of MR on the ratio of the radius to mean free path of conduction electrons with spin up of the left magnetic domain.

where \( I^\text{P(AP)} = I^\uparrow + I^\downarrow \). Then, MR is positive if the physical effect itself is negative (the resistance drops when magnetic field is applied). Now, dependence of MR on the conduction band parameters of contacting ferromagnets can be analysed. To account for a finite point contact length, we place sloping-profile domain wall inside the PC for the AP alignment of magnetizations [10]. The quantum-mechanical transmission coefficient \( D_\alpha \) through the sloping domain wall can be calculated, see for example [11]. The details can be found in article [9]. Results of magnetoresistance (MR) calculations for different ferromagnetic metals are shown in Fig. 2. Domain wall thickness between the magnetic domains is assumed to be equal to \( L = 10 \, \text{Å} \). The blue solid curve is calculated with gradient terms in the series expansion of GF’s (see Eq. (8)), and the red dashed line is calculated without gradient terms. Moreover, we investigated mean-free path effects on MR. In some cases the MR monotonously decreases as the PC cross-section increases. For some cases with a large difference in spin sub-band mean-free paths, the calculated MR shows non-monotonous behavior in the region where the radius of the contact becomes comparable with the mean-free path of electrons. We attribute this effect to the gradual change of conduction regimes from ballistic to diffusive in the vicinity of the PC upon increasing the contact cross-section size.

5. Superconducting heterostructures

Superconductivity (S) and ferromagnetism (F) are antagonistic long-range orders which cannot coexist in a homogeneous material. Superconductivity in contacts and superconductor-ferromagnet (SF) layered structures can be described by the quasiclassical theory of superconductivity of metals with impurities of a various origin. Prominent feature of ferromagnetic metals is nonequivalence of the Fermi impulses of spin-subbands of conduction bands of these metals. As a result of spin splitting of a ferromagnetic metal conduction band, singlet pair wave function becomes oscillating function of distance from the border with superconducting metal. If ferromagnetic metal is a film with a thickness, comparable with depth of penetration of pair wave function, the flow of pair wave function through the interface between superconducting and ferromagnetic metals becomes modulated with change of a thickness of ferromagnetic film because of changing conditions of an interference of the incident and reflected waves. As a result, a coupling between layers is modulated, and temperature of the superconducting transition \( T_c \) becomes nonmonotonic function of the ferromagnetic layer thickness. The oscillating and the reentrant behaviour of the \( T_c \) as functions of a ferromagnetic layer thickness were observed experimentally by our partners (Kishinev-Chernogolovka-Augsburg) in bilayers of niobium with an alloy copper/nickel. The unusual reentrant behaviour of superconductivity with double suppression of the critical temperature [12–15] was observed for the first time. At a thickness of the Nb layer of the order of 6.2 nanometers the temperature of superconducting transition \( T_c \) at first sharply fell down on increase in a thickness of ferromagnetic alloy before full suppression of
superconductivity was achieved at a thickness of the ferromagnetic alloy \( d_{\text{CuNi}} = 2.5 \) nanometers; at the further increase in a thickness \( d_{\text{CuNi}} \), the superconductivity was restored again for \( d_{\text{CuNi}} > 24 \) nanometers. Upon the further increase in a thickness of a ferromagnetic alloy the superconductivity was choked again at a thickness of an order of 38 nanometers.

The proximity effect in planar contacts between superconductors and ferromagnets was analyzed in the frame of the “dirty”-limit theory, semi-phenomenologically adapted to the case of comparable electron mean-free path and superconducting coherence length in a ferromagnet. The calculations were successfully applied to unique experiments on observation of the reentrant superconductivity in bilayers of niobium/copper-nickel alloy. The experiments on double suppression of superconductivity in the bilayers of niobium with copper-nickel alloy were correctly described. Based on material parameters, taken from the experiments, the calculations of phase diagrams for F/S/F trilayers have been done, and recommendation were formulated how to maximize the spin-valve effect in these structures.

Interference effects of the superconducting pairing wave function in thin film bilayers of Nb as a superconductor, and \( \text{Cu}_{41}\text{Ni}_{59} \) as a ferromagnetic material, lead to critical temperature oscillations and reentrant superconductivity for increasing the F-layer thickness [16–18]. The phenomenon is generated by the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) like state establishing in these geometries. So far, detailed investigations were performed on S/F bilayers. Recently, we could also realize the phenomenon in F/S bilayers where the S-metal now is grown on top of the F-material. Combining both building blocks yields an F/S/F trilayer, representing the core structure of the superconducting spin valve. Also for this geometry we observed deep critical temperature oscillations and reentrant superconductivity, which is a basis to obtain large spin-switching effect, i.e. large shift in the critical temperature, if the relative orientation of the magnetizations of the F-layers is changed from parallel to antiparallel. Ferromagnet/Superconductor/Ferromagnet (F/S/F) trilayers, in which establishing of the FFLO-like state leads to interference effects of the superconducting pairing wave function, form the core of the superconducting spin valve. The realization of strong critical temperature oscillations in such trilayers, as a function of the ferromagnetic layer thicknesses or, even more efficient, reentrant superconductivity, are the key conditions to obtain large spin valve effect, i.e. large shift in the critical temperature. Both phenomena were realized experimentally in the investigated \( \text{Cu}_{41}\text{Ni}_{59}/\text{Nb}/\text{Cu}_{41}\text{Ni}_{59} \) trilayers.

Nanolayered hybrid superconductor-ferromagnet spin-valve structure was fabricated [19,20], the resistive state of which depends on the preceding magnetic field polarity. The effect is based on a strong exchange bias (about \(-2 \) kOe) on a diluted ferromagnetic copper-nickel alloy and generation of a long-range odd-in-frequency triplet pairing component. The difference of high and low resistance states at zero magnetic field is 90% of the normal state resistance for a transport current of 250 \( \mu \)A and still around 42% for 10 \( \mu \)A. Both logic states of the structure do not require biasing fields or currents in the idle mode. F/S/F trilayers constitute a core of the superconducting spin valve. The switching effect of the spin valve is based on interference phenomena occurring due to the proximity effect at the S/F interfaces. A remarkable effect is only expected if the core structure exhibits strong critical temperature oscillations, or most favorable, reentrant superconductivity, when the thickness of the ferromagnetic layer is increased. The core structure has to be grown on an antiferromagnetic oxide layer (or such layer to be placed on top) to pin the magnetization-orientation of one of the ferromagnetic layers by exchange bias. We demonstrated that it is possible, keeping the superconducting behavior of the core structure undisturbed.
The theory of superconductor-ferromagnet heterostructures with two ferromagnetic layers predicts generation of long-range, odd-in-frequency triplet pairing at noncollinear alignment (NCA) of the magnetizations of the F layers. This triplet pairing has been detected [21] in a Nb/Cu$_{41}$Ni$_{59}$/normal conducting- (nc-) Nb/Co/CoO$_x$ spin-valve-type proximity effect heterostructure, in which a very thin Nb film between the F layers serves as a spacer of normal-conducting metal. The resistance of the sample as a function of an external magnetic field shows that for not too high fields, the system is superconducting at the collinear alignment of the Cu$_{41}$Ni$_{59}$ and Co layer magnetic moments, but switches to the normal conducting state at a NCA configuration. This indicates that the superconducting transition temperature $T_c$ for NCA is lower than the fixed measuring temperature. The existence of the minimum $T_c$ at the NCA regime below that for parallel or antiparallel alignments of the F-layers magnetic moments, is consistent with the theoretical prediction of a singlet superconductivity suppression by the long-range triplet pairing generation (see below).

The upper critical magnetic field $H_{c2}$ in thin-film ferromagnet-superconductor-ferromagnet trilayer spin-valve cores was studied experimentally and theoretically [22] in geometries perpendicular and parallel to the heterostructure surface. The series of samples with variable thicknesses $d_{F1}$ of the bottom and $d_{F2}$ of the top Cu$_{41}$Ni$_{59}$ ferromagnetic (F) layers were prepared in a single run, utilizing a wedge deposition technique. The critical field $H_{c2}$ was measured in the temperature range 0.48 K and for magnetic fields up to 9 T. A transition from oscillatory to reentrant behavior of the superconducting transition temperature versus F-layer thickness, induced by an external magnetic field, was observed for the first time. In order to properly interpret the experimental data, we developed a quasiclassical theory, enabling one to evaluate the temperature dependence of the critical field and the superconducting transition temperature for an arbitrary set of system parameters. A fairly good agreement between our experimental data and theoretical predictions was demonstrated for all samples, using a single set of fit parameters. This confirms the adequacy of the Fulde-Ferrell-Larkin-Ovchinnikov physics in determining the unusual superconducting properties of the studied Cu$_{41}$Ni$_{59}$/Nb/Cu$_{41}$Ni$_{59}$ spin-valve core trilayers.

In cooperation with the Institute of Theoretical Physics RAS (Ya.V. Fominov) and Moscow State University (M.Yu. Kupriyanov’s group) we investigated the critical temperature $T_c$ of F$_2$/F$_1$/S trilayers (Fi is a ferromagnetic metal, S is a singlet superconductor), where the long-range triplet superconducting component is generated at noncollinear magnetizations of the F layers. An asymptotically exact numerical method was employed to calculate $T_c$ as a function of the trilayer parameters, in particular, mutual orientation of magnetizations and F$_2$/F$_1$ interface transparencies. Earlier, we demonstrated [23,24] that $T_c$ in such structures can be nonmonotonic function of the angle $\alpha$ between magnetizations of the two F layers. The minimum is achieved at an intermediate $\alpha$, lying between the parallel (P, $\alpha = 0$) and antiparallel (AP, $\alpha = \pi$) cases. This implies a possibility of a triplet (TR) spin-valve effect: at temperatures above the minimum $T_c^{TR}$ but below $T_c^P$ and $T_c^{AP}$, the system is superconducting only in the vicinity of the collinear orientations. At certain configuration of parameters, we predicted a reentrant $T_c$ behaviour. At the same time, considering only the P and AP orientations, we found that both the “standard” ($T_c^P < T_c^{AP}$) and “inverse” ($T_c^P > T_c^{AP}$) switching effects are possible depending on parameters of the system (Fig. 3).

It was shown recently the existence of the anomalous dependence of the spin-triplet correlations on the angle $\alpha$ in F/F/S structures. We demonstrated a possibility of the spin-valve effect mode selection (standard switching effect, the triplet spin-valve effect or reentrant $T_c$ dependence) by the variation of the F2/F1 interface transparency (Fig. 4).
Figure 3. Critical temperature $T_c$ versus the misalignment angle $\alpha$ for the various thicknesses of the F1 layer (the F2 layer is infinitely thick) [23].

Figure 4. (Color online) Dependence of the transition temperature $T_c$ on the angle $\alpha$ between magnetizations under the different F2/F1 interface transparencies [25].

6. Summary

The quasi-classical theory of electric transport through nanoscale contacts for the case of different ferromagnetic metals has been generalized taking into account the bending of the chemical potentials near the interface. The spin-polarized conductance and MR are calculated taking into account gradient terms and covering the ballistic ($l > a$) and diffusive ($l < a$) regimes. The dependences of MR on the ratio of radius contact to mean free path of conduction electrons are shown. It can be used for interpreting the experimental data and properties of PC’s resistance of Fe-Co, Ni-Mumetall nanocontacts and tunnel structures of CoFeB/MgO/CoFe.

The works on superconductor-ferromagnet nano hybrids are reviewed in a view of the theoretical developments, as well as experimental realization of their unique superconducting properties projected on functional applications.
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