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Boundary resistance in magnetic multilayers

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Abstract

Quasiclassical boundary conditions for electrochemical potentials at the interface between diffusive ferromagnetic and non-magnetic metals are derived for the first time. An expression for the boundary resistance accurately accounts for the law of conservation of momentum as well as essential gradients of the chemical potentials. Conditions are established at which spin asymmetry of the boundary resistance has a positive or negative sign. Dependence of the spin asymmetry and the absolute value of the boundary resistance on the exchange splitting of the conduction band opens up new possibilities for estimating spin polarization of the conduction band of ferromagnetic metals. Consistency of the theory is checked on existing experimental data.

1. Introduction

The discovery of giant magnetoresistance (MR) in magnetic multilayers [1, 2], which consist of alternating layers of ferromagnetic metal (F) and normal metal (N), has stimulated intensive studies of spin-polarized transport through layered magnetic structures [3–5]. Valet and Fert [6] developed the semiclassical theory of MR in magnetic multilayers for a geometry in which the electric current is perpendicular to the layers (CPP geometry). It has been shown that when thickness of the metals in a stack is small compared with the spin diffusion length, the magnetoresistance of multilayers can be calculated within the two-current series-resistor model [7–9]. In this case MR is expressed via the resistance of the F/N interface (boundary resistance), $r_{\uparrow(\downarrow)} = 2[1 \mp \gamma_{F/N}]R^*_{F/N}$, and the bulk specific resistance of the ferromagnetic layer, $\rho_{\uparrow(\downarrow)} = 2[1 \mp \beta_F]\rho^*_F$ [6]. In these expressions $\gamma_{F/N}$ and β_F are parameters of spin asymmetry of the boundary and bulk scattering resistances. It follows from [6, 10] that their linear combination, $\beta_{F1}\rho^*_{F1}t_{F1} + \gamma_1 R^*_{F/N}$, determines sign of MR in layered $[F1/N/F2/N] \times n$ structures (t_F is the thickness of the ferromagnetic layers). In [11, 12] positive (inverse) MR due to negative value of $\gamma_{F/N}$ in (F1/Cr/F2/Cr) multilayers was observed for the first time.

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Theoretical calculations of the boundary resistance [13–15] established the strong influence of the spin-dependent band structure of ferromagnetic metals on magnetoresistance. Using the approach of [13] and transmission probabilities through the F/N interface, calculated earlier from first principles [16], Stiles and Penn [14] obtained from numerical calculations a negative value of $\gamma_{F/N}$ for the Fe/Cr interface and a positive $\gamma_{F/N}$ for iron-group ferromagnet–noble metal interfaces. However, conditions on the parameters of contacting materials resulting in negative or positive values of spin asymmetry of the boundary resistance were not discussed in the above papers.

In this paper we derive for the first time quasiclassical boundary conditions for electrochemical potentials of diffusive ferromagnetic and normal metals, which can be used for solution of a wide class of problems in spintronics. Our expression for the boundary resistance accurately accounts for the law of conservation of momentum as well as essential gradients of the chemical potentials. We establish conditions on parameters of the contacting metals, at which spin asymmetry of the boundary resistance has a positive or negative sign. Dependence of the spin asymmetry and the boundary resistance on the exchange splitting of the conduction band offers one more way to estimate spin polarization of conduction band of ferromagnetic metals. We give an example of such an estimation.

2. Boundary conditions for electrochemical potentials

We derive boundary conditions for electrochemical potentials of diffusive metals using the quasiclassical Green functions technique. A 'diffusive ferromagnetic metal' is a metal in which spin splitting of the conduction band is small compared with the momentum relaxation rate for conduction electrons. Let us consider that axis x is perpendicular to the F/N boundary, and neglect reversal of the electron spin upon transmission through the interface. Then, for each of the metals, equations for the Green functions $g_\alpha(\mathbf{n}, x, \rho, t)$ read [17]:

$$v_{x,\alpha}\frac{\partial g_{a,\alpha}}{\partial x} + \mathbf{v}_\parallel \frac{\partial g_{s,\alpha}}{\partial \rho} + \frac{1}{\tau_\alpha}\left(g_{s,\alpha} - \overline{g}_{s,\alpha}\right) = 0, \tag{1}$$

$$v_{x,\alpha}\frac{\partial g_{s,\alpha}}{\partial x} + \mathbf{v}_\parallel \frac{\partial g_{a,\alpha}}{\partial \rho} + \frac{g_{a,\alpha}}{\tau_\alpha} = 0. \tag{2}$$

Here $\mathbf{n} = \mathbf{p}_{x,\alpha}/|\mathbf{p}_\alpha|$; $g_{s(a),\alpha} = (1/2)[g_\alpha(n_x, x, \rho, t) \pm g_\alpha(-n_x, x, \rho, t)]$ is the single-particle quasiclassical Green function symmetric (antisymmetric) with respect to a projection of the Fermi momentum $\mathbf{p}_{x,\alpha}$ on the axis x, v_x is a projection of the Fermi velocity on the axis x, $\alpha = (\uparrow, \downarrow)$ is a spin index and $\rho = (y, z)$ is a coordinate in a plane of the contact. The bar above $g_{s,\alpha}$ means integration over the solid angle: $\overline{g}_{s,\alpha} = \oint \mathrm{d}\Omega/2\pi\, g_{s,\alpha}$.

The boundary conditions to equations (1) and (2) are as follows [17]:

$$g^{\mathrm{F}}_{a,\alpha}(0) = g^{\mathrm{N}}_{a,\alpha}(0) = \begin{cases} g_{a,\alpha}(0), & p_\parallel < p^{\mathrm{F}}_\alpha, p^{\mathrm{N}} \\ 0, & \min(p^{\mathrm{F}}_\alpha, p^{\mathrm{N}}) < p_\parallel, \end{cases} \tag{3}$$

$$2R_\alpha g_{a,\alpha}(0) = D_\alpha\left(g^{\mathrm{F}}_{s,\alpha}(0) - g^{\mathrm{N}}_{s,\alpha}(0)\right). \tag{4}$$

In equations (3) and (4) p^{F}_α and p^{N} are the Fermi momenta in ferromagnetic and normal metals, respectively, $p_\parallel$ is a projection of a momentum on the plane of the contact, D_α and $R_\alpha = 1 - D_\alpha$ are the spin-dependent, quantum-mechanical transmission and reflection coefficients. Boundary conditions (3) and (4) obey the specular reflection law:

$$p_\parallel = p^{\mathrm{F}}_\downarrow \sin\theta_\downarrow = p^{\mathrm{F}}_\uparrow \sin\theta_\uparrow = p^{\mathrm{N}} \sin\theta_N. \tag{5}$$

The angles θ in (5) are measured from the axis x, a range of variation for the biggest one is $[0, \pi/2]$. The quasiclassical equations (1) and (2), and the boundary conditions (3) and (4) are formulated for a single electron trajectory determined by the angles φ and θ.

Upon solution of the system of equations (1) and (2) we shall consider that the ferromagnet is located to the left of the boundary $x = 0$, and the normal metal to the right ($x > 0$), and that the functions $g_{s,\alpha}$ are homogeneous in the plane of the contact. Then, the system of equations (1) and (2) can be solved in the form of integral equations for the functions $g_{a,\alpha}$ and $g_{s,\alpha}$ in the energy representation, $g_{s,\alpha}(\varepsilon) = 2\tanh(\varepsilon/2T) + f_{s,\alpha}(\varepsilon)$:

$$f^{\mathrm{N}}_{s,\alpha}(x) = g^{\mathrm{N}}_{a,\alpha}(x) + \frac{1}{l_{x,\alpha}}\int_x^\infty \mathrm{d}\xi\, \mathrm{e}^{\frac{x-\xi}{l_{x,\alpha}}} \overline{f}^{\mathrm{N}}_{s,\alpha}(\xi), \tag{6}$$

$$f^{\mathrm{F}}_{s,\alpha}(x) = -g^{\mathrm{F}}_{a,\alpha}(x) + \frac{1}{l_{x,\alpha}}\int_{-\infty}^x \mathrm{d}\xi\, \mathrm{e}^{\frac{\xi-x}{l_{x,\alpha}}} \overline{f}^{\mathrm{F}}_{s,\alpha}(\xi). \tag{7}$$

In a dirty metal the solid-angle averaged function $\overline{f}_{s,\alpha}(\xi)$ obeys the diffusion equation with a decay length which is much longer than the mean free path l_α. Then, we expand $\overline{f}^{\mathrm{F(N)}}_{s,\alpha}(\xi)$ in the right-hand side of equations (6) and (7) near point x and take out from the integrals terms independent of ξ. Substituting the resulting expansions into the boundary condition (4) we find:

$$2g_{a,\alpha}(0) = D_\alpha\left[\left(1 - l^{\mathrm{F}}_{x,\alpha}\frac{\mathrm{d}}{\mathrm{d}x}\right)\overline{f}^{\mathrm{F}}_{s,\alpha}(x) - \left(1 + l^{\mathrm{N}}_{x,\alpha}\frac{\mathrm{d}}{\mathrm{d}x}\right)\overline{f}^{\mathrm{N}}_{s,\alpha}(x)\right]_{x=0}. \tag{8}$$

To formulate boundary conditions for the functions $\overline{f}^{\mathrm{F(N)}}_{s,\alpha}$ (which are, in fact, chemical potentials; see below) we use a matching procedure proposed in [18]. From equation (1) it follows that for distances of the order of $l_{x,\alpha}$ from the interface

$$\overline{l_{x,\alpha}\frac{\mathrm{d}g_{a,\alpha}}{\mathrm{d}x}} = 0. \tag{9}$$

Hence

$$\overline{l_{x,\alpha}g_{a,\alpha}} = C = \text{constant} \tag{10}$$

in each of the metals. Now we calculate, for example, C^{F} using the expression (8) for $g_{a,\alpha}(x = 0)$. Then we calculate C^{F} far from the interface using an approximate expression for $g^{\mathrm{F}}_{a,\alpha}(x)$,

$$g^{\mathrm{F}}_{a,\alpha}(x) = -l^{\mathrm{F}}_{x,\alpha}\frac{\mathrm{d}\overline{f}^{\mathrm{F}}_{s,\alpha}(x)}{\mathrm{d}x}, \tag{11}$$

which follows from equation (2) after expansion of $g^{\mathrm{F}}_{a,\alpha}$ on Legendre polynomials. Equating values of the constant C^{F} calculated in the two ways, and applying the relationship between the averaged Green function and the electrochemical potential, $\overline{f}_{s,\alpha} = (2/\pi)\mu_\alpha$, we receive the boundary condition for the electrochemical potentials at the interface $x = 0$:

$$l^{\mathrm{F}}_\alpha\frac{\mathrm{d}\mu^{\mathrm{F}}_\alpha(0)}{\mathrm{d}x} = \delta_\alpha\left(\mu^{\mathrm{N}}_\alpha(0) - \mu^{\mathrm{F}}_\alpha(0)\right), \tag{12}$$

where

$$\begin{aligned}
&\delta_\alpha = \frac{\delta_{1,\alpha}}{1-\delta_{2,\alpha}}, \qquad \delta_{1,\alpha} = \frac{3}{2}\int\frac{\mathrm{d}\Omega_{F,\alpha}}{2\pi}\cos(\theta_{F,\alpha})D_\alpha,\\
&\delta_{2,\alpha} = \frac{3}{2}\int\frac{\mathrm{d}\Omega_{F,\alpha}}{2\pi}\left[x + \left(\frac{p^{\mathrm{F}}_\alpha}{p^{\mathrm{N}}}\right)^2\cos(\theta_N)\right]x D_\alpha,\\
&x = \cos(\theta_{F,\alpha}), \qquad \mathrm{d}\Omega_{F,\alpha} = \sin(\theta_{F,\alpha})\,\mathrm{d}\theta_{F,\alpha}\,\mathrm{d}\varphi.
\end{aligned} \tag{13}$$

The limits of angular integration must satisfy specular reflection conditions at the interface, equation (5). When deriving equation (12) we have used conservation of the current density at the interface, which follows from equation (3),

$$j_\alpha^{\mathrm{F}}(0) = \frac{\sigma_\alpha^{\mathrm{F}}}{e}\frac{\mathrm{d}\mu_\alpha^{\mathrm{F}}(0)}{\mathrm{d}x} = \frac{\sigma_\alpha^{\mathrm{N}}}{e}\frac{\mathrm{d}\mu_\alpha^{\mathrm{N}}(0)}{\mathrm{d}x} = j_\alpha^{\mathrm{N}}(0), \tag{14}$$

where $\sigma_\alpha^{\mathrm{F}}$ and $\sigma_\alpha^{\mathrm{N}}$ are the bulk, spin-channel conductivities of the metals:

$$\sigma_\alpha^{\mathrm{F(N)}} = \frac{e^2(p_\alpha^{\mathrm{F(N)}})^2 l_\alpha^{\mathrm{F(N)}}}{6\pi^2}. \tag{15}$$

Equation (14) is actually the second, complementary to equation (12), boundary condition for the semiclassical description of the spin-polarized transport in magnetic multilayers in terms of electrochemical potential.

3. Resistance of the interface

The derivative from the electrochemical potential in equation (12) can be expressed in terms of the current density (14), and we find the spin-dependent resistance of the interface r_α:

$$\mu_\alpha^{\mathrm{N}}(0) - \mu_\alpha^{\mathrm{F}}(0) = e r_\alpha\, j_\alpha, \tag{16}$$

$$r_\alpha = \frac{6\pi^2}{e^2(p_\alpha^{\mathrm{F}})^2 A}\frac{1-\delta_{2,\alpha}}{\delta_{1,\alpha}}, \tag{17}$$

where A is the area of the contact. It follows from equation (17) that, in the quasiclassical approach at specular reflection from the interface, the boundary resistance between ferromagnetic and normal metals is determined only by the Fermi momenta of the contacting metals and the coefficient of transmission through the interface.

Experimental data are given for the spin asymmetry of boundary resistance, $\gamma_{\mathrm{F/N}}$, and for the renormalized resistance of the interface, $AR^*_{\mathrm{F/N}}$, determined as follows:

$$\gamma_{\mathrm{F/N}} = \frac{r_\downarrow - r_\uparrow}{r_\uparrow + r_\downarrow}, \qquad AR^*_{\mathrm{F/N}} = \frac{A}{4}(r_\downarrow + r_\uparrow). \tag{18}$$

To calculate dependence of $\gamma_{\mathrm{F/N}}$ and $AR^*_{\mathrm{F/N}}$ on the Fermi momentum of the non-magnetic metal for various values of the ferromagnet conduction band polarization we have used the Fermi-momentum-mismatch model for the transmission coefficient: $D_\alpha = 4v_{x,\alpha}^{\mathrm{N}} v_{x,\alpha}^{\mathrm{F}}/[(v_{x,\alpha}^{\mathrm{N}})^2 + (v_{x,\alpha}^{\mathrm{F}})^2]$. Results are presented in figures 1 ($p_\uparrow^{\mathrm{F}} > p_\downarrow^{\mathrm{F}} > p^{\mathrm{N}}$), 2 ($p_\uparrow^{\mathrm{F}} > p^{\mathrm{N}} > p_\downarrow^{\mathrm{F}}$) and 3 ($p^{\mathrm{N}} > p_\uparrow^{\mathrm{F}} > p_\downarrow^{\mathrm{F}}$). From our calculations it follows that for a non-magnetic metal with a low density of conduction electrons (small value of the Fermi momentum p^{N}) the spin asymmetry of the boundary resistance $\gamma_{\mathrm{F/N}}$ is always negative (figure 1). On the contrary, for a non-magnetic metal with a high density of conduction electrons the spin asymmetry of the boundary resistance is always positive (figure 3). In an intermediate situation $\gamma_{\mathrm{F/N}}$ can be negative as well as positive (see figure 2). To attain the maximum amplitude of negative magnetoresistance in F/N multilayers the spin asymmetry of the boundary resistance $\gamma_{\mathrm{F/N}}$ and the asymmetry of the bulk resistance β_{F} should be both positive and close to unity. According to our calculations, the Fermi momentum of the non-magnetic metal should be as close as possible to the Fermi momentum of the majority subband of the ferromagnetic metal (figure 2, $p^{\mathrm{N}}/p_\uparrow^{\mathrm{F}} \to 1.0$ and figure 3, $p_\uparrow^{\mathrm{F}}/p^{\mathrm{N}} \to 1.0$). The spin asymmetry of the bulk resistance can be adjusted by the type and concentration of impurities in the ferromagnetic metal [19]. Clearly, similar arguments can be applied to the opposite case of negative values of $\gamma_{\mathrm{F/N}}$ and β_{F}, which will result in positive magnetoresistance in multilayers of alternating

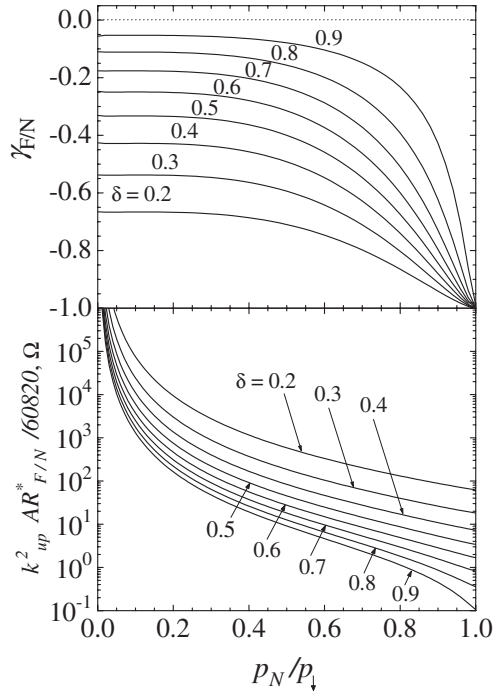

Figure 1. Dependence of the spin asymmetry of boundary resistance $\gamma_{F/N}$ (top field) and the renormalized resistance of the interface $AR^*_{F/N}$ (bottom field) on the Fermi momentum of the non-magnetic metal for the case ($p^F_\uparrow > p^F_\downarrow > p^N$).

ferromagnetic and non-magnetic metals. However, a negative asymmetry of the bulk resistance is met less often [19]. Competition of opposite sign asymmetries of boundary and bulk resistances can result in negative or positive magnetoresistance depending on the choice of materials and thickness of the ferromagnetic layers [12].

4. Discussion of experiments

Experiments on CPP transport in multilayers are very complicated because the resistance of a stack of layers of nanometre thickness is very small (of the order of fΩ m^2). Nevertheless, available experimental data (see reviews [4, 20] and references in them, and also [12, 21, 22]) allow us to test the internal consistency of the theory. Multilayers of the iron-group ferromagnetic metals with noble metals most likely belong to case 3 ($p^N > p^F_\uparrow > p^F_\downarrow$), and the parameter of the spin asymmetry $\gamma_{F/N}$ is positive [4, 12, 20–22]. For example, $\gamma_{Co/Cu} \simeq 0.77$ [4]. Then, intersection of the horizontal dashed line $\gamma_{Co/Cu} \simeq 0.77$ in the top field of figure 3 with the curve $\gamma_{F/N}(p^N)$, corresponding to $\delta = 0.6$, gives $p^F_\uparrow/p^N \simeq 0.7$. Accepting $p^F_\uparrow = 1.0$ Å^{-1} as a trial value for the Fermi momentum of the majority subband of cobalt we get $p^{Cu} \simeq 1.41$ Å^{-1}, which is a fairly good fit for the free-electron-model value for copper, $p^{Cu}_{FEM} \simeq 1.36$ Å^{-1} [23]. There are data for silver as the non-magnetic spacer: $\gamma_{Co/Ag} \simeq 0.85$ [22]. In a similar way, we obtain from figure 3 $p^{Ag} \simeq 1.22$ Å^{-1}, which fits well the free-electron-model value $p^{Ag}_{FEM} \simeq 1.20$ Å^{-1} [23].

Let us look now at consistency of the theory with boundary resistance data. For the combination Co/Cu, $AR^*_{Co/Cu}(\text{exp}) \simeq 0.51$ fΩ m^2 [4]. Continuing the vertical dashed line

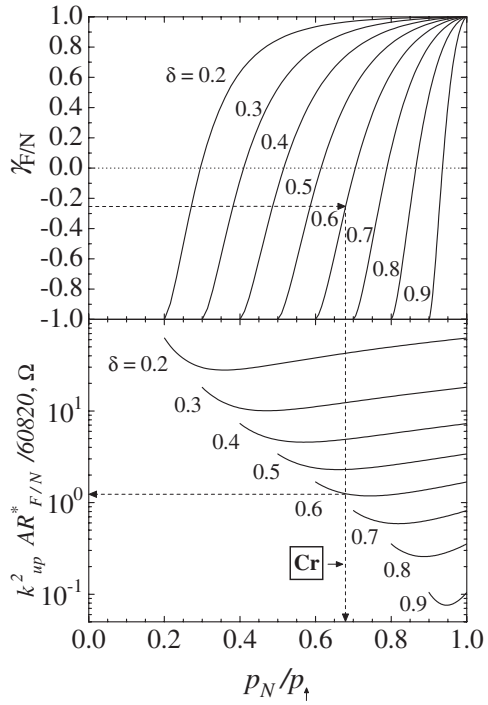

Figure 2. The same as in figure 1, but for the case ($p^{\mathrm{F}}_{\uparrow} > p^{\mathrm{N}} > p^{\mathrm{F}}_{\downarrow}$).

for copper in figure 3 into the bottom field until intersection with the curve $AR^{*}_{\mathrm{F/N}}(p^{\mathrm{N}})$, corresponding to $\delta = 0.6$, we obtain $AR^{*}_{\mathrm{Co/Cu}}(\mathrm{theor}) \simeq 0.74$ fΩ m^2. A similar procedure for silver gives $AR^{*}_{\mathrm{Co/Ag}}(\mathrm{theor}) \simeq 0.69$ fΩ m^2 (compare with $AR^{*}_{\mathrm{Co/Ag}}(\mathrm{exp}) \simeq 0.56$ fΩ m^2 [12, 22]). It worth noting that the theory reproduces fairly well the closeness of the boundary resistances of the Co/Cu and Co/Ag interfaces.

For Co/Cr multilayers the asymmetry of the boundary resistance is negative, $\gamma_{\mathrm{Co/Cr}} \simeq -0.24$ [12]. This value is admissible for cases 1 (figure 1, top field) and 2 (figure 2, top field). Without details, we conclude that the first case does not match the expected value of the conduction band polarization parameter for Co $\delta \sim 0.6 \pm 0.1$ as well as results for boundary resistances about two orders of magnitude higher. The second case (see dashed lines in figure 2) results in a Fermi momentum $p^{\mathrm{Cr}} \sim 0.68$ Å^{-1}, and in a boundary resistance $AR^{*}_{\mathrm{Co/Cr}}(\mathrm{theor}) \simeq 0.61$ fΩ m^2. The Fermi momentum is satisfactory in the frame of the free electron model [23]. The boundary resistance is close enough to the experimental value $AR^{*}_{\mathrm{Co/Cr}}(\mathrm{exp}) \simeq 0.48$ fΩ m^2 [12]. We expect that better matching of the band structures of cobalt and chromium, both belonging to the iron-group metals, would result in a weaker influence of the real band structure on the boundary resistance.

A discrepancy with experiment in an absolute value of boundary resistance of about 20–45% does not seem catastrophic for the following reasons. First, the trial choice of $p^{\mathrm{F}}_{\uparrow} = 1.0$ Å^{-1} was not optimized. Second, we used the free s-electron model as a background for the theory. One might expect that for an interface between a metal with predominantly d-electron conduction band (iron group) and an s-electron metal (Cu, Ag), reduced overlapping and symmetry mismatch may noticeably increase the boundary resistance. Third, García and

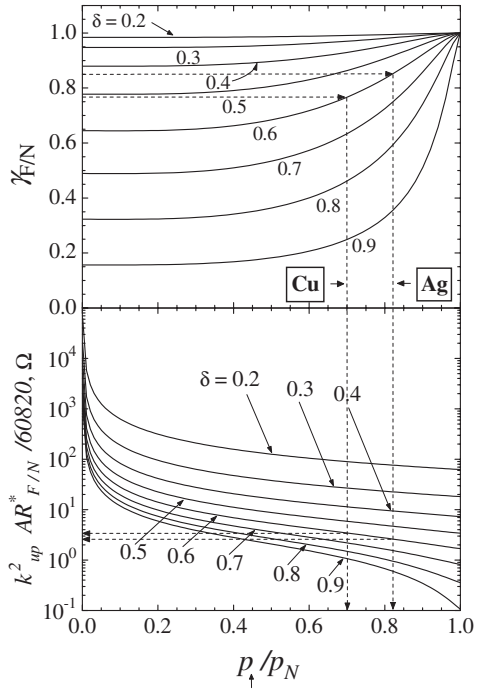

Figure 3. The same as in figure 1, but for the case ($p^{\mathrm{N}} > p^{\mathrm{F}}_{\uparrow} > p^{\mathrm{F}}_{\downarrow}$).

Stoll [24] have shown (figure 2 of [24]) that the interface roughness also increases boundary resistance. This increase is estimated to be below 20–60% for different models and magnitudes of the interface roughness and reasonable differences in the Fermi momenta of contacting metals. The interface roughness is much less important for the case of CPP transport, considered in this paper, than for current-in-plane (CIP) transport [25]. Finally, spin reversal at the interface opposes both the previously considered processes, decreasing the boundary resistance. The reversal of the electron spin by the spin–orbit interaction is always expected upon refraction of the electron wave or scattering on roughness at the interface between two metals. Quantitative analysis of the competition between the above minor mechanisms of boundary resistance is beyond the scope of this paper. However, it is our expectation that the key quantity, spin asymmetry of the boundary resistance $\gamma_{\mathrm{F/N}}$, is only weakly dependent on band structure matching, interface roughness, spin reversal, etc, because of considerable cancellations in the dimensionless ratio (equation (18)).

Our trial evaluations show that the experimental data for the spin asymmetry of boundary resistance and the absolute value of boundary resistance in Co/Cu, Co/Ag and Co/Cr multilayers can be consistently described with the use of the spin polarization parameter for the conduction band of cobalt $\delta \simeq 0.6$. At the level of the experimental accuracy and completeness of the theory the estimated value of δ is identical to $\delta \simeq 0.57$, which we have estimated [26] from experiments of García *et al* [27] on magnetoresistance of cobalt nanocontacts. Experiments on Andreev spectroscopy cite similar values of δ for cobalt [28]. Thus, the spin asymmetry of the boundary resistance in combination with the absolute value of the boundary resistance can be used for estimations of the spin-polarization parameter δ of the conduction band of ferromagnetic metals.

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