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Parametric spin-valve effect in superconductor/ferromagnet structures

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The equations of the microscopic theory of superconductivity in the diffusion limit (Usadel equations) are used to interpret experimental data from a study of the spin-valve effect in epitaxial Nb/Ho structures. The cause of the quasimetallic behavior of their critical dependences is determined. The influence of the triplet component of the superconducting condensate on the critical temperature is studied. *Published by AIP Publishing.* [<http://dx.doi.org/10.1063/1.4965895>]

1. Introduction

The special properties of superconducting (S)/ferromagnetic (F) structures which are superconducting because of the proximity effect originate in the exchange interaction field that determines the oscillatory character of the damping of the ordering parameter in the F -layer.^{1–3} As a result, the superconducting condensate can be found in 0- and π -states in S/F/S and its spectrum of states^{4,5} in multilayer structures shows up in the oscillatory character of the critical dependences.^{1–3} Because of these properties, S/F structures offer promise in superconducting nanoelectronics.⁶ The combination of effects observed during changes in the magnetic state of S/F structures is the basis of their applications in superconducting spintronics.³

Spin valves (SV) based on three-layer F1/S/F2 structures in which the superconducting state is controlled by “ P - AP ” SV inversions^{7,8} or by rotation (a triplet SV⁹) of the magnetic moment of one of the F layers are an elementary spintronic device. The resulting ΔT_c effect can be substantial for S -layer thicknesses d_S close to critical, $d_{S,cr}$, and can approach T_c for a narrow range of F -layer thicknesses^{7–12} (ΔT_c is the difference between the critical temperatures T_{cAP} and T_{cP} of the states with antiparallel and parallel magnetic moments, respectively, or in a second case, between the maximum and minimum critical temperatures T_c , and $d_{S,cr}$ is the thickness at which superconductivity vanishes^{13–15}). These strict conditions are fully consistent with experiment: $\Delta T_c = 30 \text{ mK} \ll T_c$ for structures with weak ferromagnetism¹⁶ and ranges from -50 mK to 40 mK for structures with strong ferromagnetism^{3,16}). A marked SV effect is possible in structures with high quality S-F interfaces^{16–19} that are fully determined by the material parameters of the systems;²⁰ for structures with strong ferromagnets, here the predicted result correlates with calculations outside the diffusion approximation.^{21–23} The SV effect occurs in a fundamentally simpler way in multilayer structures, especially S/F1/N/F2 (N is a normal metal) and this explains their advantage in applications.^{11,24–27} On the other hand, the high sensitivity of ΔT_c to perturbations in d_S – $d_{S,sc}$ means that an elementary SV can be regarded as an “instrument” for

solving some fundamental problems. In this paper we analyze the following results of a recent experiment on Ho(d_0)/Nb/Ho(d_F) structures²⁸ from this very standpoint:

- (1) “quasi-S/N” the behavior of $T_{cP}(d_F)$, $T_{cAP}(d_F)$;
- (2) strong P - AP effect, $\Delta T_c = 400 \text{ mK}$. Although this value of ΔT_c is an order of magnitude higher than those obtained for F1/S/F2 structures, result (1) is more significant, since it provides a direct indication of processes taking place at the S-F contact.

2. Material parameters and critical characteristics of Nb/Ho systems

Fitting of the $T_c(d_F)$ dependences measured for an Ho(d_0)/Nb/Ho(d_F) structure with magnetic moments in the F -layers that were collinear to the specified in-plane direction²⁸ in an $d_S \ll \xi_S$ approximation based on the Usadel equations²⁹ yielded a very small value for the effective exchange energy parameter, $E_{ex} \sim 10 \text{ K}$ and coherence lengths $\xi_S \approx 32 \text{ nm}$ and $\xi_F \approx 30 \text{ nm}$ ($\xi_{S(F)} = \sqrt{D_{S(F)}/2\pi T_S}$, D_S and D_F are the diffusion coefficients, and T_S is the critical temperature for the bulk superconductor). This value of ξ_F corresponds to the decay length for the experimental $T_c(d_F)$ curve and is typical of S/N structures (for the Nb/Cu system the coherence length in the normal metal is $\xi_N \sim 35 \text{ nm}$ (Refs. 30 and 31)) and the unusual value of ξ_S for niobium appears in the fitting procedure as a factor for suppression of the critical temperature as $E_{ex} \rightarrow 0$ (the usual values for niobium are $\xi_S \sim 6$ – 7 nm (Refs. 5, 20, 30, and 31)).

Measurements of the magnetic properties of this structure²⁸ do not confirm the results of the fitting procedure: the in-plane saturation magnetization M_s and the residual magnetization M_r were 2500 and 2100 G, respectively, or roughly 20%–30% less than the maximum $M_{s,m} \sim 3100$ – 3400 G obtained if the magnetic moment is directed along the easy axis of magnetization.³² Since the value of $M_{s,m}$ corresponds to an exchange energy of $\sim 10^4 \text{ K}$,³³ in this case it is difficult to speak of a mechanism leading to almost complete elimination of the effect of the exchange field.

We now estimate E_{ex} and the other material parameters of this Nb/Ho system based on the available measurements.^{28,34} In terms of the Usadel equations, an S/F system is characterized by the parameters E_{ex} , ξ_S , ξ_F , and T_S , as well as by the ratio $p = \rho_S/\rho_F$ of the normal low-temperature resistivities of the S- and F-layers (or $\gamma = p\xi_S/\xi_F$ (Ref. 35)) and the quantum-mechanical transparency coefficient of the S-F interface, \mathcal{T} (or the parameter γ_b (Ref. 35)). ξ_S can be determined from measurements of the upper perpendicular critical field for a series of F/S(d_S)/F structures; in this case it is unknown. The effective temperatures T_S is most reliably determined from the asymptotic behavior of the $T_c(d_S)$ characteristic. This characteristic has been measured for a series of Nb(d_S)/Ho(12 nm) structures³⁴ fabricated under roughly the same conditions as the Ho(d_0)/Nb/Ho(d_F) series.²⁸ p can be estimated from the published^{28,34} resistivities $\rho_S = 3.6 \mu\Omega \text{ cm}$ of a 30-nm-thick Nb film and $\rho_F = 95 \mu\Omega \text{ cm}$ of a 100-nm-thick Ho film. Since niobium films with thicknesses of 15–30 nm are used in the experiments, the measured values can be treated as lower boundaries for ρ_S . On the other hand, for holmium the resistivity was determined in the asymptotic region although an Ho film of thickness 12 nm is used in the experiment to measure the $T_c(d_S)$ characteristic. This means (given the data of Ref. 33) that the possible values of p can lie within a rather wide neighborhood of $p \sim 0.05$. For the transparency coefficient we have an upper bound $\mathcal{T} \leq 0.5$, found from the Fermi velocities $v_{F,Nb} = 0.27 \cdot 10^6 \text{ m/s}$ (Ref. 36) and $v_{F,Ho} = 1.6 \cdot 10^6 \text{ m/s}$.²⁸ A reliable estimate of the parameters E_{ex} , ξ_F , and \mathcal{T} requires simultaneous fitting of the characteristics $T_c(d_S)$ and $T_c(d_F)$.²⁰ Here, however, this is impossible since the series of two- and three-layered structures differ, at least, in their exchange energies. In fact, for two-layered structures we can assume that E_{ex} is close to the maximum, since the measured values of $M_S \approx M_r \approx 3000 \text{ G}$ are close to $M_{S,m}$.²⁸

All the parameters of the system are, therefore, undetermined to some extent. However, in order to find the cause of the quasi-S/N behavior of the $T_c(d_F)$ characteristic, it is sufficient to analyze the $T_c(d_S)$ characteristic, since in this case it provides more definite information on the parameters. In fact, fitting the curves to the exact solution of the Usadel equations^{4,5} yields the following:

- (1) For arbitrary $E_{ex} \geq 50 \text{ K}$, in the parameter space ξ_S , ξ_F , p , \mathcal{T} there is a set at each point of which the theoretical curve will reproduce the experimental dependence with the same error. Up to an exchange energy of $E_{ex} \sim (3\text{--}4) \times 10^3 \text{ K}$ there is a region of values of these parameters in which the conditions for the diffusion limit are met.
- (2) The set of permissible values of the system parameters is bounded by the condition $\xi_S, \xi_F < 20 \text{ nm}$.

Some examples of $T_c(d_S)$, curves calculated for different values of the parameters are shown in Fig. 1.

The existence of a set of fit parameters implies that the criterion for a parametric degeneracy of the $T_c(d_S)$ characteristic is satisfied: the thicknesses of the layers in the structure exceed the corresponding correlation lengths.^{20,30,31} This is also confirmed by the fact that the dependences calculated in a single mode approximation reproduce the experimental curve equally well.

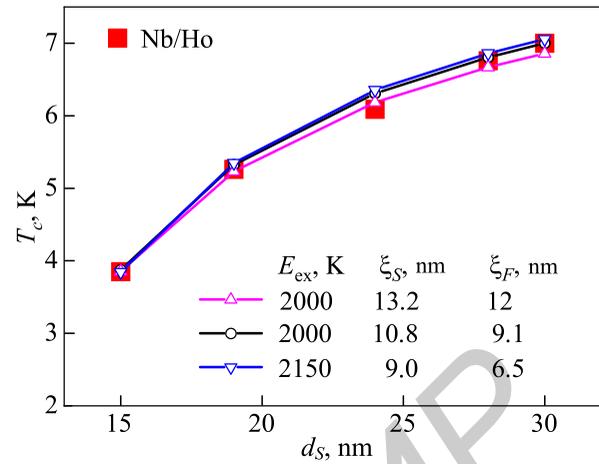


Fig. 1. Experimental (points)³⁴ and calculated (curves with open symbols) critical temperatures of an Nb(d_S)/Ho(12 nm) structure.

3. Quasi-S/N behavior of the critical characteristics of the Nb/Ho system

We now consider the fact that the characteristic $T_c(d_F)$ has been measured for a very wide range of thicknesses (this series included samples with thicknesses $d_0 = 10 \text{ nm}$, $d_S = 20 \text{ nm}$, and thicknesses d_F of 10, 40, and 70 nm (Ref. 28)). In this case, the mean free path ℓ_F and the low-temperature resistivity $\rho_F \sim 1/\ell_F$ can vary from sample to sample (thus, $\xi_F, \gamma_b \sim \ell_F^{1/2}$ change),³⁷ which may have a significant effect on the critical temperature.³⁸ How strongly the material parameters vary can be judged on the basis of the available measurements for Nb and Pd₈₁Nd₁₉ films.³⁹ When the thickness is increased from 10 to 70 nm, the resistivity $\rho(d)$ of the samples decreases by roughly a factor of 3.3 and 2.5, respectively, and is very well approximated by the formula

$$\rho(d) = \rho_a \left[1 + \frac{3\ell_a}{2d} \left(E_3 \left(\frac{d}{\ell_a} \right) - E_5 \left(\frac{d}{\ell_a} \right) - \frac{1}{4} \right) \right]^{-1}, \quad (1)$$

where $E_n(x)$ is the exponential integral,⁴⁰ and ρ_a, ℓ_a are the asymptotic values of the functions $\rho(d), \ell(d)$. (The value of the parameter ℓ_a for these metals turns out to be unphysical; thus, Eq. (1) must be treated as empirical here.)

Given these considerations, we now calculate $T_{cP}(d_F)$ and $T_{cAP}(d_F)$ for $d_S = 30 \text{ nm}$ (close to the critical value) with one of the sets of parameters obtained by fitting the $T_c(d_S)$ curves: $E_{ex} = 2000 \text{ K}$, $p = 0.05$, $\mathcal{T} = 0.5$, $T_S = 9.2 \text{ K}$, $\xi_S = 10.8 \text{ nm}$, and $\xi_F = 9.1 \text{ nm}$. We assume that $p(d_F) = \rho_S/\rho_F(d_F)$ (12 nm) = 0.05 and $p_a = 0.09$; then $\ell_a = 21 \text{ nm}$. The result of the calculation is shown in Fig. 2, which also shows $T_{cP}(d_F)$ and $T_{cAP}(d_F)$ calculated for the same parameters but with fixed p , along with a segment of the critical temperature as a function of the thickness of the N-layer, d_N , of an Nb/Cu/Nb S/N structure.

Figure 2 shows that the calculated $T_{cP}(d_F)$ and $T_{cAP}(d_F)$, for an Nb/Ho system neglecting drift in the resistivity of the F-layer are typical of S/F structures with a strong ferromagnet. Once the drift is taken into account, the shape of the curves changes and becomes similar to the characteristic for an Nb/Cu system. We can also see that the magnitude of the spin-valve effect increases as the thickness of the F-layer is raised.

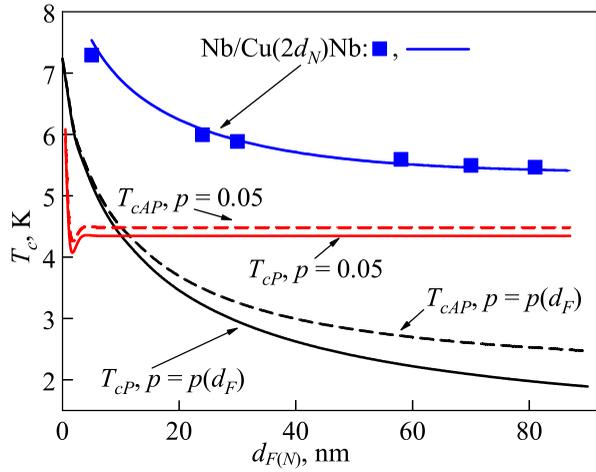


Fig. 2. Critical temperatures of an $F(d_{0,F})/S/F(d_F)$ structure calculated for parallel and antiparallel magnetic moments of the F -layers with and without drift in the parameters (curves). The measured (points) and calculated critical temperatures of an Nb/Cu/Nb structure³⁰ are also shown.

4. The state function of the superconducting condensate in an F0/S/F structure

The matrix solution of the one-dimensional linearized Usadel equations is written in terms of the vector state functions $\Phi(z) = \Phi_+(z) \oplus \Phi_-(z)$, where the column vectors $\Phi_+(z)$ and $\Phi_-(z)$ are formed, respectively, by frequency-even $\Phi_{\omega,+}(z)$ and frequency-odd $\Phi_{\omega,-}(z)$ components of the anomalous Green functions^{4,5} (ω are the Matsubara frequencies, the Z axis is perpendicular to the layers, and XY coordinate plane is coincident with the symmetry plane of the S -layer). The vector function $\Phi_+(z)$ describes the singlet component of the condensate (s_0), and the vector function $\Phi_-(z)$, the triplet (s_{10}) component corresponding to spin projection 0. These calculations show that for the system studied here, the characteristics, $T_c(d_S)$ and $T_c(d_F)$ obtained from an exact solution of the equations are also reproduced in a single-mode approximation with a small offset of the fit parameters. In the single-mode approximation, the system is characterized by the two-component vector function of state $\Phi(z) = \Phi_+(z) \oplus \Phi_-(z)$.

We now calculate $\Phi(z)$ with $d_0 = 10$ nm and $d_S = 20$ nm for several values of d_F in the range of 10–70 nm, moving along the $T_{cP}(d_F)$ and $T_{cAP}(d_F)$ curves. (Here the system parameters were chosen using the condition that the theoretical points coincide with four of the six experimental points.) We first separate the “pure effect” of the drift in the parameter p , assuming that the length ζ_F is independent of thickness. The result is shown in Fig. 3. (Part of the region of the F -layer is shown, since, as can be seen in Fig. 3, the state function vanishes for all d_F at distances less than 10 nm from the interface.) The picture does not change fundamentally if we include the drift in all the parameters that depend on the mean free path. This can be seen in Fig. 4, which shows plots of the s_{10} component of the state function (Figs. 4(b) and 4(c)) and of part of the s_0 component in a small neighborhood of the S-F interface (Fig. 4(a)), calculated with the drift in p and ζ_F taken into account. (As in the previous example, the functions $\Phi_+(z)$ for the P and AP magnetic states are almost identical, so only one graph is shown.)

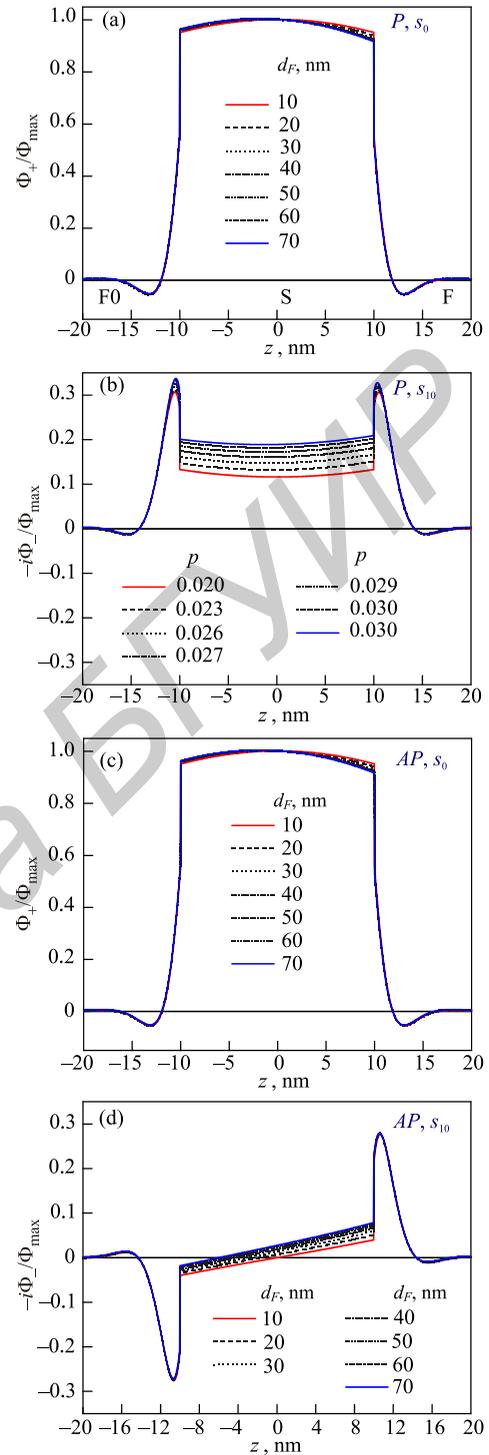


Fig. 3. Singlet (s_0) and triplet (s_{10}) components of the state function of the condensate in an F0/S/F structure in P and AP magnetic states with the drift in the parameter p taken into account.

5. Discussion of results

As the above remarks imply, the quasi-S/N behavior of the $T_c(d_F)$ characteristics of the Nb/Ho system are caused, first, by drift of the system parameters as the thickness of the F -layer is increased, second, by the high exchange energy, and, third, by the high resistivity of holmium and the low transparency of the S-F interface.

Because of the high exchange energy, the superconducting condensate penetrates into the F -layer to a small depth determined by the characteristic length $\zeta_F = \zeta_F \sqrt{2\pi T_S/E_{ex}}$, so that the ordering parameter rapidly approaches its

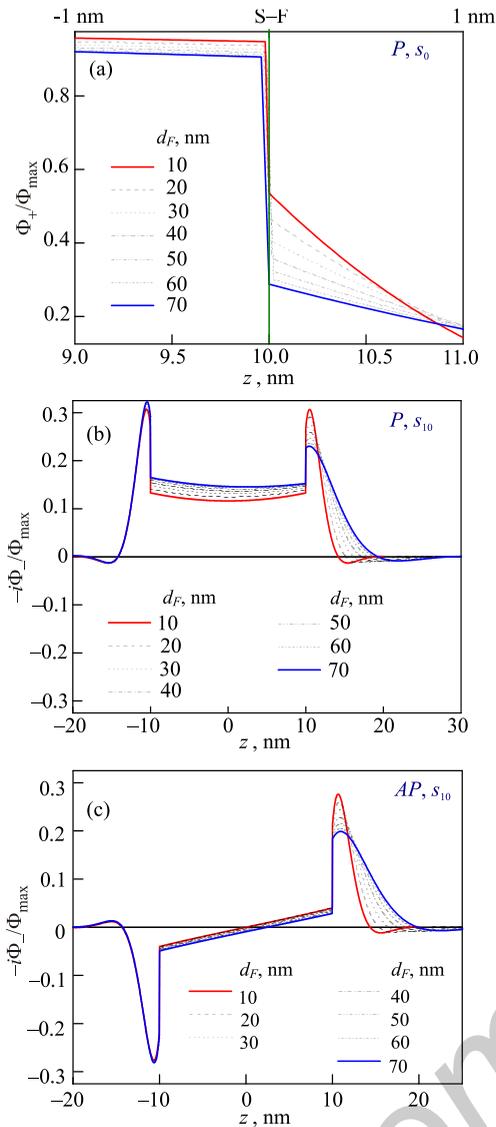


Fig. 4. Singlet (s_0) and triplet (s_{10}) components of the state function of the condensate in an F0/S/F structure in P and AP magnetic states constructed with the drift in the parameters p and ζ_F taken into account.

asymptotic dependence as the thickness of the F -layer is increased (Fig. 3). Thus, each value of T_c measured for $d_F \gg \zeta_F$ is essentially asymptotic, but it does drift because of the increased rates of electron pair diffusion through the S-F interface owing to the reduction in the resistivity ρ_F (Fig. 2). The large values of $\rho_F(d_F)$ (throughout the entire range of variation) and the low transparency of the S-F interface mean that superconductivity is weakly suppressed in the S -layer (Figs. 3(a) and 4(a)) and the critical temperatures of the structure are high; this explains the observability of the effect.

The experiments,²⁸ therefore, were a direct measurement of the dependence of the critical temperature and the magnitude of the SV effect on the parameter that determines the rate of diffusion through the S-F interface. In order to obtain more complete information on this process, let us examine the “evolution” of the state function with varying d_F (i.e., ρ_F). Figures 3(a), 3(b), 4(a), and 4(b), reflect the direct dependence of the critical temperature of an F0/S/F structure with a thick S layer and almost semi-infinite F -layers in the P state on the relationship between the s_{10} - and s_0 -

components of the superconducting condensate and may illustrate the result of processes leading to the generation of triplet pairs and the destruction of singlet pairs as the diffusion rate increases. Furthermore, in Figs. 3(a), 3(c), and 4(a), we can see that the singlet component is almost insensitive to inversion of the magnetic moments of the F layers. The triplet state, on the other hand, rapidly changes the configuration. First, during $P \rightarrow AP$ transitions it reaches zero in the S -layer (a general property, valid for any number of double layers). In particular, for a symmetric F/S/F structure the function $\Phi_-(z)$ transforms from symmetric to antisymmetric (this is easily proved by the matrix method¹⁴). Second, the fraction of the triplet component in the S -layer for the AP state is less than for the P state. Third, as Figs. 3(d) and 4(c) show, the triplet component in the AP state is much more stable with respect to perturbations at the S-F interface. This explains the gradual divergence of the $T_{cP}(d_F)$ and $T_{cAP}(d_F)$ characteristics with increasing d_F (or ρ).

6. Conclusion

An analysis of experimental data on the spin-valve effect in epitaxial Nb/Ho structures based on an exact solution of the Usadel equations shows the following: the observed “quasi-S/N” dependences of the critical temperature on the thickness of the ferromagnetic layer, as well as the magnitude of the effect, are caused by drift in the parameters of the structure as the thickness of the ferromagnetic layer is varied. Calculations of the state function have been used to study the influence of the generation of the triple component of the superconducting condensate on the critical temperature. For a detailed quantitative analysis of the effect of processes at the S-F boundary, the experiments on structures in the Nb/Ho system will have to be supplemented by appropriate measurements.

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