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Author(s): Silaev, Mikhail

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Large enhancement of spin pumping due to the surface bound states in normal metal–superconductor structures

M. A. Silaev

*Department of Physics and Nanoscience Center, University of Jyväskylä, P.O. Box 35 (YFL), FI-40014 University of Jyväskylä, Finland;
Moscow Institute of Physics and Technology, Dolgoprudny 141700, Russia;
and Institute for Physics of Microstructures, Russian Academy of Sciences, 603950 Nizhny Novgorod, GSP-105, Russia*



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We show that the spin pumping from ferromagnetic insulator into the adjacent metallic spin sink can be strongly stimulated by the superconducting correlations. The key physical mechanism responsible for this effect is the presence of Andreev bound states at the ferromagnetic insulator/superconductor interface. We consider the minimal model when these states appear because of the suppressed pairing constant within the interfacial normal layer. For thin normal layers we obtain a strongly peaked temperature dependence of the Gilbert damping coefficient which has been recently observed in such systems. For thicker normal layers the Gilbert damping monotonically increases down to the temperatures much smaller than the critical one. The suggested model paves the way to controlling the temperature dependence of the spin pumping by fabricating hybrid normal metal–superconductor spin sinks.

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Introduction. Spin transport and spin dynamics in superconductors have attracted significant attention recently [1–7]. Quite interesting experimental results have been obtained for the spin pumping effects [8–18] which in general play the central role in spintronics [19–21].

It was found that superconducting correlations can lead either to the significant suppression [8,22] or to the significant enhancement [9–13,17,22] of the Gilbert damping (GD) coefficient in systems consisting of superconducting and ferromagnetic layers, such as in the generic example shown in Fig. 1. The basic mechanism for changing GD in such systems is the spin pumping effect [21]. This mechanism is based on the spin angular momentum transfer from the ferromagnet into the adjacent metallic film via the pumped spin current $\mathbf{i}(t)$ generated by the time-dependent magnetization $\mathbf{m}(t)$.

The spin relaxation in the metallic spin sink leads to the dampinglike spin torque and modifies the effective GD coefficient of the system. In this way the suppression of GD with decreasing temperature $T < T_c$ in systems with superconducting spin sink [8] can be qualitatively understood as resulting from the freezing out of quasiparticles in the superconductor [23]. However, the strong increase of GD with lowering temperature [9–13,17] seems to be counterintuitive and its understanding requires further theoretical efforts.

In ferromagnetic insulator (FI)/superconductor (S) bilayers GdN/NbN the peaked behavior of GD as a function of temperature has been observed [13]. The maximal GD reached at about $T \approx 0.7T_c$ is several times larger than in the normal state $\delta\alpha/\delta\alpha_N \sim 2-3$, where $\delta\alpha$ is the spin-pumping related change of GD.

Because of several reasons such behavior cannot be explained [24] by the coherence peak of spin susceptibility in homogeneous superconductors [25]. First, this peak oc-

curs at higher temperature $T \approx 0.9T_c$ and it is strongly suppressed by increasing the Dynes parameter [26] Γ . For NbN where the realistic values are [27,28] $\Gamma/T_c \approx 0.1-0.2$ the maximal GD enhancement due to the coherence peak is [24] $\max(\delta\alpha)/\delta\alpha_N \sim 0.2-0.3$. Such behavior is typical for the linewidths of nuclear magnetic resonance [29,30] and electronic paramagnetic resonance [31] in superconductors. It is clearly different from the observed behavior of GD in FI/S systems [13] which have an order of magnitude larger peak $\delta\alpha/\delta\alpha_N \sim 2-3$ at significantly lower temperatures $T \approx 0.7T_c$.

In this Rapid Communication we suggest a minimal theoretical model which explains the large enhancement of GD in FI/S structures. The key physical mechanism responsible for this effect is the existence of Andreev bound states localized at the FI/S interface. Such states appear due to the suppressed pairing within the interfacial normal layer [32–35] (N) as illustrated in Fig. 1. Quasiparticles become localized near superconducting order parameter inhomogeneities in result of Andreev reflections converting electrons to holes and vice versa. Shown at the top of Fig. 1 are the spatial profiles of the order parameter $\Delta(x)$ and the local density of states (DOS) $N(x)$ at the subgap energy $\varepsilon = 0.5\Delta_0$, where Δ_0 is the bulk energy gap at $T = 0$. The overall N/S film thickness is $d_S = 3\xi_0$, where $\xi_0 = \sqrt{D_S/T_{c0}}$ is the coherence length, D_S is the diffusion constant in S, and T_{c0} is the bulk critical temperature. Near the interface at $x = 0$ the DOS is enhanced due to the subgap quasiparticle states which are formed in the N/S structure [36–39], and occupy the certain energy interval below the bulk gap. The existence of surface bound states in N/S structures is demonstrated [40] in Figs. 2(a) and 2(c) where the $N(x, \varepsilon)$ profiles are shown to have a maximum at $x = 0$ and energies which depend on d_N . The order parameter

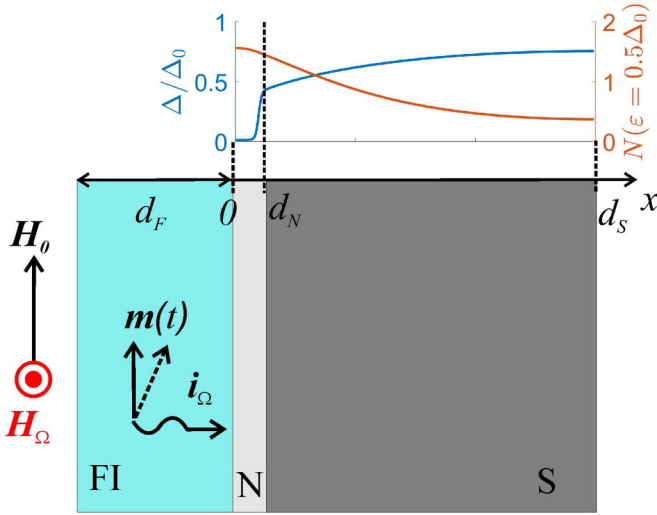


FIG. 1. Schematic setup of the ferromagnetic insulator (FI) film with the adjacent metallic spin sink consisting of normal (N) and superconducting (S) layers. The constant external magnetic field is $H_0\mathbf{x}$. The magnetization precession $\mathbf{m}(t)$ is driven by the external magnetic field $H_0 e^{i\Omega t}\mathbf{y}$. It generates spin current i_Ω pumped from FI to the spin sink. Upper panel shows the coordinate dependencies of the order parameter $\Delta(x)$ and local density of states $N(x)$ at the energy $\varepsilon = 0.5\Delta_0$ for $d_N = 0.2\xi_0$, $d_S = 3\xi_0$, $T = 0.7T_c$.

and DOS in Figs. 1 and 2 are calculated within the Usadel theory [41] as explained below. In Fig. 1 we choose identical diffusion coefficients in N and S layers $D_N = D_S = D$ while in Fig. 2 $D_N = 0.05D_S$.

At low frequencies $\Omega \ll \Delta_0$ the DOS enhancement leads to the increased probability of the magnon absorption by conductivity electrons in the N/S layer. Qualitatively, at a given energy level this probability is determined by the number of available states for transition $N(\varepsilon)N(\varepsilon + \Omega) \approx N^2(\varepsilon)$ and the difference of occupation numbers $n_0(\varepsilon + \Omega) - n_0(\varepsilon) \approx \Omega \partial_\varepsilon n_0$ where $n_0(\varepsilon) = \tanh(\varepsilon/2T)$ is the equilibrium distribution function. The product of these factors leads to the energy-resolved magnon absorption probability $P_m = \Omega N^2 \partial_\varepsilon n_0$.

In Figs. 2(b) and 2(d) one can see that $P_m(\varepsilon)$ at $T = 0.7T_{c0}$ is enhanced at the boundary of N layer $x = 0$ (red curves) as compared to $x = d_S$ (blue curves). As we show by an exact calculation below, this mechanism leads to the large enhancement of spin pumping in the N/S films. Interestingly, besides explaining the large peak of the spin pumping for $d_N \approx 0.2\xi_0$ the model described above yields also the qualitatively different regime with almost monotonic increase of GD down to the temperatures $T \ll T_c$. This behavior is obtained for $d_N \gg \xi_0 \sqrt{D_N/D_S}$ when the bound states are pushed down to lower energies as shown in Fig. 2(c). As a result the absorption probability is enhanced for quasiparticles with $\varepsilon \ll \Delta_0$ which are not frozen out down to the significantly low temperatures determined by the Thouless energy $\varepsilon_{th} \approx D_N/d_N^2 \ll T_c$. Similar behavior of GD has been observed experimentally in Py/Nb/Pt superconducting heterostructures [12,17], although its physical origin can be different.

Model of spin pumping. To quantify the spin pumping effect we consider the microscopic model of the spin-dependent

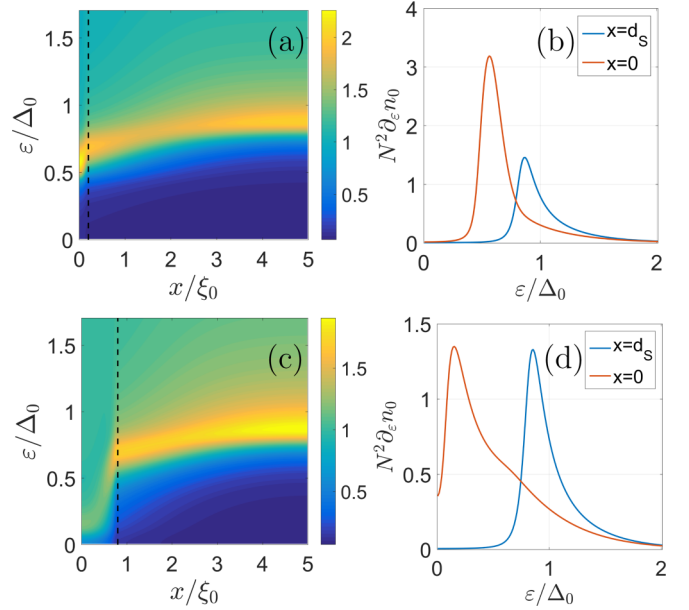


FIG. 2. (a),(c) Density of states profile $N(\varepsilon, x)$ in the N/S structure. The position of the N/S boundary shown by the dashed line is at (a) $d_N = 0.2\xi_0 \approx \sqrt{D_N/T_{c0}}$ and (c) $d_N = 0.8\xi_0 \approx 4\sqrt{D_N/T_{c0}}$. $T = 0.7T_{c0}$, $\Gamma = 0.1T_{c0}$, $d_S = 5\xi_0$, $D_N = 0.05D_S$. Plots for other d_S are shown in the Supplemental material [40]. (b),(d) Magnon absorption probability $P_m(\varepsilon) = \Omega \partial_\varepsilon n_0 N^2$ for the frequency $\Omega = 0.02T_{c0}$. Red and blue curves are taken at $x = 0$ and $x = d_S$, respectively. Parameters are the same as in (a) and (b).

scattering of electrons at the FI interface [40,42,43]. As we show below, it formally yields the spin current identical to the one given by the interfacial exchange interaction between the localized spins in FI and conduction electrons in the adjacent metal [44,45]. Within this model the local spin polarization close to the interface $\mathbf{S}(t)$ acts as an effective field for the localized magnetic moments. This process can be taken into account by introducing the additional term $\mathbf{i}(t)$ into the Landau-Lifshitz-Gilbert equation

$$(1 + \alpha \mathbf{m} \times) \partial_t \mathbf{m} + \gamma \mathbf{m} \times \mathbf{H}_{\text{eff}} = \mathbf{i}/S_{F0} d_F, \quad (1)$$

$$\mathbf{i}(t) = J_{sd} \mathbf{S}(t) \times \mathbf{m}(t). \quad (2)$$

Here S_{F0} is the equilibrium spin density in F , d_F is the F film thickness, \mathbf{H}_{eff} is the effective field, and α is the intrinsic Gilbert damping coefficient. The term $\mathbf{i}(t)$ can be interpreted as the spin current between FI and metal.

To calculate $\mathbf{S}(t)$ we need to find the spin response of the superconductor to the interfacial exchange field. In the linear regime it is given by

$$\mathbf{S}_\Omega = \nu h_{\text{eff}} \chi_m \mathbf{m}_\Omega, \quad (3)$$

where we introduce the effective exchange field $h_{\text{eff}} = J_{sd}/d_S$, normal metal DOS at the Fermi level ν , and the local spin susceptibility χ_m .

The spin-pumping related change of the GD is determined by the dissipative part of the susceptibility

$$\delta\alpha = CT_{c0} \text{Im} \chi_m / \Omega, \quad (4)$$

where the dimensionless coefficient determining the coupling strength between the FI and metallic films is [24]

$$C = \frac{h_{\text{eff}}}{T_{c0}} \frac{v h_{\text{eff}}}{S_{F0}} \frac{d_S}{d_F}. \quad (5)$$

This coefficient determines the overall amplitude of the GD change (4) due to the spin pumping. It drops out, however, from the relative change $\delta\alpha/\delta\alpha_N$ determined by the superconducting correlation.

Calculation of the time-dependent spin response. What is left is to calculate the local spin susceptibility χ_m in the Eq. (4) for the FI/N/S structure in Fig. 1. We do so by developing the microscopic kinetic theory of spin pumping generalizing the quasiclassical approach [2,43,46,47] to the time-dependent situation.

The magnetization of conduction electrons is determined by spin accumulation and can be written in terms of the Keldysh quasiclassical Green's function (GF) as

$$S(t) = -\nu \text{Tr} [\hat{\tau}_3 \hat{\sigma} g^K(t, t)]/8, \quad (6)$$

where g^K is the (2×2) matrix Keldysh component of the quasiclassical GF matrix $\check{g} = \begin{pmatrix} \hat{g}_0^R & \hat{g}_0^K \\ 0 & \hat{g}_0^A \end{pmatrix}$ which depends on two times and a single spatial coordinate variable $\check{g} = \check{g}(t_1, t_2, \mathbf{r})$.

GF \check{g} obeys the Usadel equation

$$\{\hat{\tau}_3 \partial_t, \check{g}\}_t + \nabla(D\check{g} \circ \nabla\check{g}) = \Delta[\hat{\tau}_1, \check{g}] + [\hat{\Gamma}, \check{g}] - [\check{\Sigma}_{so}, \check{g}]_t, \quad (7)$$

where $\hat{\sigma}_k$, $\hat{\tau}_k$, $k = 0, 1, 2, 3$ are Pauli matrices and D is the diffusion coefficient.

The commutator operator is defined as $[X, g]_t = X(t_1)g(t_1, t_2) - g(t_1, t_2)X(t_2)$, similarly for anticommutator $\{, \}_t$. The symbolic product operator is given by $(A \circ B)(t_1, t_2) = \int dt A(t_1, t)B(t, t_2)$.

Spin relaxation is determined by the spin-orbital scattering self-energy

$$\check{\Sigma}_{so} = \sigma \cdot \hat{g}\sigma/(6\tau_{so}). \quad (8)$$

The self-consistency equation for the gap function is

$$\Delta = \lambda \text{Tr}[\hat{\tau}_1 \hat{g}^K]/4, \quad (9)$$

where λ is the pairing coefficient. In our model we assume the pairing constant to be suppressed in the N region $\lambda(x < d_N) = 0.05\lambda(x > d_N)$ as compared to its value in S. We scan over the values of the diffusion coefficient in the N layer D_N while keeping it fixed in the S layer D_S . The inelastic scattering is described by the Dynes [26] parameter which enters into Eq. (7) as the matrix in Nambu-Keldysh space with $\hat{\Gamma}^{R,A} = \pm\Gamma\hat{\tau}_3$. This term describes both the DOS singularity broadening and the relaxation of nonequilibrium distribution functions as described below.

Equation (7) is supplemented by the *dynamical boundary conditions* at $x = 0$ describing the spin splitting and pumping induced by the electron scattering at the FI interface with time-dependent magnetization. These boundary conditions are derived [40] from the spin-dependent scattering matrix \hat{S} connecting the incident $\hat{\psi}_i$ and reflected $\hat{\psi}_r$ electronic waves $\hat{\psi}_r = \hat{S}(t)\hat{\psi}_i$. For frequencies small compared to the exchange field in FI we use the adiabatic approximation which yields the expression $\hat{S} = e^{i(m\hat{\sigma})\hat{\tau}_3\Theta/2}$, where Θ is the time-independent

spin-mixing angle. Then, assume that $|\Theta| \ll 1$ and

$$D\check{g} \circ \partial_x \check{g}(x = 0) = iJ_{sd}[\sigma\mathbf{m}\hat{\tau}_3, \check{g}]_t, \quad (10)$$

where $\mathbf{m} = \mathbf{m}(t)$ is the time-dependent magnetization. Within the minimal band model of the FI [42,43] the interfacial exchange constant is expressed through the spin-mixing angle as $J_{sd} = \frac{\nu v_F}{4} \int_{-1}^1 d\hat{p}_x |\hat{p}_x| \Theta(\hat{p}_x)$, where \hat{p}_x is the electron momentum projection on the interface normal. Equation (10) generalizes the static boundary condition at the spin-active interface [42,43,47,48] to the case of time-dependent magnetization. The induced spin current is obtained using the general expression $\mathbf{i}(t) = \pi\nu D \text{Tr}[\hat{\sigma}\check{g} \circ \partial_x \check{g}](t, t)$. With the help of Eqs. (10) and (6) it yields the phenomenological equation (2).

Introducing the usual parametrization of quasiclassical Keldysh function in terms of the distribution function $\hat{g}^K = \hat{g}^R \circ \hat{f} - \hat{f} \circ \hat{g}^A$ we can identify the terms which are essential to calculate linear response in the low-frequency limit. Expanding the energy representation of \hat{g}^K to the first order in Ω we obtain the nonequilibrium correction

$$\delta\hat{g}^K = (\hat{\sigma}\mathbf{m}\Omega) \left[(\hat{g}_0^R - \hat{g}_0^A)f_h + \frac{\Omega\partial_\varepsilon n_0}{2}(\hat{g}_h^R + \hat{g}_h^A) \right], \quad (11)$$

where we parametrize the spin-dependent corrections as follows: $\hat{f} = (\hat{\sigma}\mathbf{m}\Omega)f_h$ and $\delta\hat{g}^{R,A} = (\hat{\sigma}\mathbf{m}\Omega)\delta\hat{g}_h^{R,A}$. In contrast to stationary nonequilibrium situations [46] when only the first term in (11) is important, the time-dependent case requires taking into account also the second term with the corrections of spectral functions [24]. This is different from previous calculations of spin dynamics [49,50] which neglected correction to spectral functions and obtained nonphysical zero-temperature dissipation in the superconducting state.

In the low-frequency limit the calculation is simplified by neglecting the frequency dependence of the perturbed spectral GF in (11).

Using (11) we write the time-dependent spin polarization in the metallic film as follows:

$$S_\Omega = i\Omega\mathbf{m}\Omega \int_{-\infty}^{\infty} d\varepsilon [2Nf_h + (\hat{g}_{3h}^R + \hat{g}_{3h}^A)\partial_\varepsilon n_0], \quad (12)$$

where $N = \text{Tr}(\hat{\tau}_3 \hat{g}^R)/2$ is the local DOS and $\hat{g}_{3h}^{R,A} = \text{Tr}(\hat{\tau}_3 \hat{g}_h^{R,A})/2$. Equations for zero-order spectral function $\hat{g}_0^{R,A}(\varepsilon, x)$, corrections $\hat{g}_h^{R,A}(\varepsilon, x)$, and the distribution function $f_h(\varepsilon, \Omega, x)$ are obtained straightforwardly [40] from Eqs. (7) and (10). The zero-order GF $\hat{g}_0^{R,A}(\varepsilon, x)$ are calculated in the N/S structure self-consistently together with the order parameter (9). This gives in particular the $\Delta(x)$ and $N(\varepsilon, x)$ profiles shown in Figs. 1 and 2. The corrections f_h and $\hat{g}_h^{R,A}$ are determined by the linear equation [40].

Results and discussion. Using the described formalism we calculate the nonequilibrium spin polarization (12) in the N/S structure shown in Fig. 1. This gives us the local susceptibility (3) and the excess GD (4). The resulting temperature dependencies of $\delta\alpha(T)$ are shown in Fig. 3 for various parameters. The first column in Fig. 3 corresponds to $\Gamma = 0.1T_{c0}$ and identical diffusion coefficients in N and S layers. In the absence of N layer $d_N = 0$ there is a usual coherence peak at $T \approx 0.9T_c$ with the small amplitude $\delta\alpha/\delta\alpha_N \approx 1.4$. Adding the thin N layer with $d_N > 0.1\xi_0$ leads to the increase of the peak amplitude to $\delta\alpha/\delta\alpha_N \approx 1.9$ and shifting to lower temperatures.

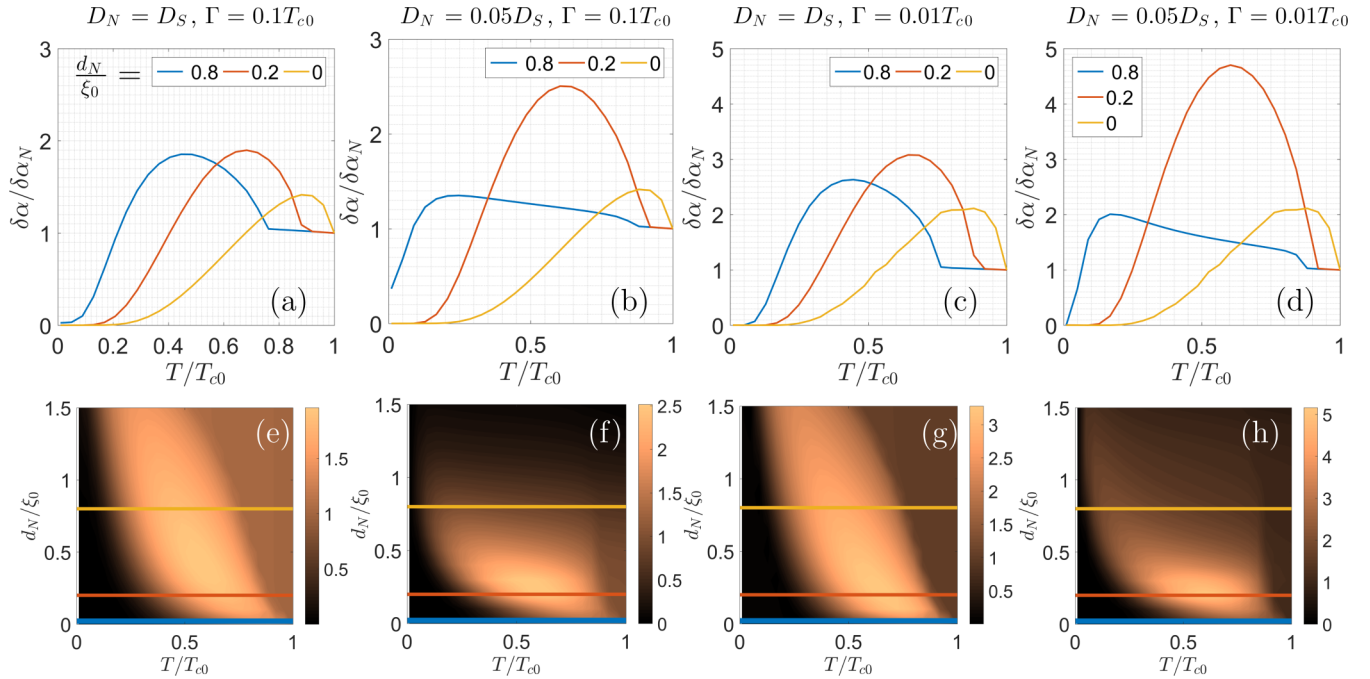


FIG. 3. Upper row: temperature dependencies of the GD $\delta\alpha(T)$ in FI/N/S systems. The three curves in each plot correspond to $d_N/\xi_0 = 0.8, 0.2$, and 0 . Lower row: color plots of the functions $\delta\alpha(d_N, T)/\delta\alpha_N$. Horizontal lines in each panel are positioned as a guide for the eyes at $d_N/\xi_0 = 0.8, 0.2$, and 0 corresponding to the curves in the upper plot. The four columns correspond to various Dynes parameters $\Gamma/T_{c0} = 0.1, 0.01$ and ratios of diffusion coefficients in N and S layers $D_N/D_S = 1, 0.05$ specified on top of the panels. Common parameters are $d_S = 3\xi_0, \tau_{sn}T_{c0} = 1, \Omega = 0.02T_{c0}$.

The peak is enhanced by decreasing the diffusion coefficient D_N in the normal layer. Qualitatively, this leads to better localization of surface bound states and hence to the increase of surface DOS. As shown in the second column of Fig. 3 for $D_N = 0.05D_S$ and $\Gamma = 0.1T_{c0}$ the peak is enhanced to $\delta\alpha/\delta\alpha_N \approx 2.5$ reached at $T \approx 0.7T_c$ with $d_N = 0.2\xi_0$. This behavior is quite similar to the experimental observation [13]. For larger $d_N > 0.5\xi_0$ the temperature dependence is qualitatively changed to the monotonic increase down to the low temperatures. As shown by the yellow curve with $d_N = 0.8\xi_0$ the increase continues to $T \approx 0.1T_c$.

An even larger increase is obtained for smaller Dynes parameters $\Gamma = 0.01T_{c0}$ as shown in the third and fourth columns of Fig. 3. For $D_N = D_S$ we obtain the maximal value $\delta\alpha/\delta\alpha_N = 3$. For $D_N = 0.05D_S$ we obtain the maximal value $\delta\alpha/\delta\alpha_N = 4.8$. For all values of Γ we note that for $d_N \gg \sqrt{D_N/D_S}\xi_0$ the monotonically increasing $\delta\alpha(T)$ is obtained down to the threshold temperature of the order of Thouless energy $\varepsilon_{th} \approx D_N/d_N^2$. As one can see in the color plots in Figs. 3(f) and 3(h), for increasing d_N it can be rather small, $\varepsilon_{th} \ll T_c$.

The introduced model can explain the observed spin-pumping enhancement in the GdN/NbN system [13] assuming that there is a naturally formed thin normal layer at the FI/S interface. The pairing suppression at the interface can result from various reasons, including magnetic disorder [51,52], strong usual disorder [53] or the band structure modification [54]. It is straightforward to check our prediction of the enhanced GD by fabricating artificial FI/N/S structures with various parameters.

The behavior of $\delta\alpha(T)$ obtained in Figs. 3(b) and 3(d) with $d_N = 0.8\xi_0$ is qualitatively similar to that observed experimentally in Py/Nb/Pt heterostructures [12,17]. In the equilibrium state of our model the spin-triplet superconductivity is absent. Therefore the monotonic increase of GD due to the superconducting correlations is not in principle an exclusive feature of the system with spin supercurrents. However, the spin-triplet correlations are generated in the nonequilibrium case (11) providing [24] significant contribution to the spin response (12).

The developed quasiclassical theory of spin pumping can be generalized to the case of metallic ferromagnets by introducing the finite spin-dependent tunneling probability through the F/S interface [47,55,56] to the boundary condition (10). This provides the way to study charge and heat transport induced by the magnetic precession as well as spin torques induced by voltage and temperature biases [57–61].

Conclusions. We have developed the general formalism to calculate spin pumping in spatially inhomogeneous metallic films with spin-active interfaces. As an example we have considered the FI/N/S structure and found that the presence of quasiparticle bound states localized near the spin-active interface provides strong enhancement of spin pumping which shows up in the strong increase of the GD coefficient with decreasing temperature below T_c . The model explains the large peak of GD in Gd/NbN structures and shows the way to controlling spin-pumping properties in superconducting systems. In the clean case quasiparticle bound states in such system lead to the geometrical resonances of spin pumping [62]. It would be interesting

to study how spin pumping can be used for probing various other bound states in superconductors including vortex core states [63,64] and surface states in unconventional superconductors [22,65,66].

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