Spin-flip Enhanced Thermoelectricity in Superconductor-Ferromagnet Bilayers

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Abstract. We study the effects of Zeeman-splitting and spin-flip scattering in a superconductor (S) on the thermoelectric properties of a tunneling contact to a metallic ferromagnet (F) using the Green's function method. A giant thermopower has been theoretically predicted and experimentally observed in such structures. This huge thermoelectric effect is attributed to the spin-dependent particle-hole asymmetry in the tunneling density of states in the S/F heterostructure. Here, we evaluate the S density of states and thermopower for a range of temperatures, Zeeman-splitting, and spin-flip scattering. In contrast to the naive expectation, we find that the spin-flip scattering strongly enhances the thermoelectric performance of the system in the low-field and low-temperature regime. This is attributed to a complex interplay between the charge and spin conductances caused by the softening of the spin-dependent superconducting gaps. The maximal value of the thermopower exceeds k_B/e by a factor of ~ 5 and has a nonmonotonic dependence on Zeeman-splitting and spin-flip rate. We also demonstrate that the incoherent broadening leads to a drastic reduction of the thermoelectric performance.

1. Introduction

The field of thermoelectrics has drawn an enormous amount of interest in recent years driven by the goal of recovering unused heat and converting it into usable electricity. The thermoelectric (TE) or Seebeck effect refers to the conversion of temperature differences (ΔT) across a system into usable electrical potential (V). The effect is quantified in terms of the so-called Seebeck coefficient S as $S = -V/\Delta T$. The efficiency of a TE device is determined by the type of materials used in making the device. Thus, a major focus of efforts towards addressing today's energy challenge is to find more efficient TE structures [1].

In a conductor, an applied temperature gradient drives the flow of electrons and holes. These two contributions are in general unequal due to varying density of states (DOS) giving rise to a net charge current, or equivalently a net voltage in an open circuit called thermopower. However, the variation in DOS is very small and the thermoelectric response is negligibly small. Hence, achieving a strong variation in the DOS near the Fermi energy is the key to enhancing TE effects in a system. A superconductor with Zeeman-splitting breaks the spin-dependent electron-hole symmetry and its interface with a ferromagnetic metal results in spin-polarized conductance leading to a large Seebeck effect. Following this principle, large spin-dependent TE effects arising as a result of coupled spin and heat transports were theoretically proposed [2–10] and experimentally demonstrated [11–13] in spin-polarized superconducting tunnel contacts. Spin transport in S/F structures with large Zeeman-splitting of the DOS [14], and TE effects of a quantum dot coupled to ferromagnetic and superconducting electrodes (F-quantum dot-S) [15, 16] has also been studied. It has been shown that the combination of spin-polarized tunneling at the ferromagnetic-quantum dot interface and the utilization of an external magnetic field that Zeeman-splits the dot energy level leads to large values of the thermopower [16]. Most the Zeeman-split superconducting tunnel heterostructures exhibiting large TE effects require a large externally applied magnetic field [11, 17]. A recent experiment [12] reported observation of large TE effects employing the spin splitting field provided by proximity coupling with a ferromagnetic insulator, instead of an applied magnetic field.

Spin-flip scattering, in general, has a detrimental effect on superconductivity. It reduces the gap and may lead to gapless superconductivity. It has been disregarded in several studies of TE effects in S/F bilayers. Recent works investigate the influence of spin-flip mechanism on the critical current [18] and critical temperature [18, 19], the DOS [20], the spatial and energy dependences of the anomalous Green's function [21], and quasiparticles distribution and the superconducting state [22] in a spin-split S, of S/F multilayered systems.

In this work, we predict a low-field spin-flip induced enhancement of the Seebeck coefficient in an S/F bilayer. We incorporate spin-flip scattering and incoherent broadening self-consistently within the quasiclassical description of S [23, 24]. The spin-flip scattering shifts the peaks of the DOS toward zero energy [see Fig. 2] thus reducing the gap. We find that spin-flip scattering suppresses (enhances) the Seebeck coefficient at large (low) spin splitting of the DOS. Overall, the maximal Seebeck coefficient is relatively insensitive to the spin-flip rate as long as the system is away from the gapless superconductivity regime. This counterintuitive effect results from a complex interplay between the total and spin-polarized DOS instigated by spin-flip mediated softening of the gap. The Seebeck coefficient consists of a ratio of an integral of the total DOS to an integral of the spin-polarized DOS. We further find that incoherent broadening reduces the thermoelectric performance of the system consistent with expectations.

This paper is structured as follows. In section 2, we outline the model and theoretical framework employed to describe the TE effects in an S/F bilayer. Section 3 presents the dependence of the Seebeck coefficient on the spin-flip scattering rate and incoherent broadening. We conclude with a discussion and a summary in section 4.

2. Model

We consider a ferromagnet layer coupled via a spin-polarized tunneling contact to a thin spin-split superconductor layer, i.e., the thickness of the superconducting film is smaller than the coherence length. A sketch of the device under consideration has been shown in figure 1. The spin splitting field is orientated collinear to the magnetization direction of the ferromagnet film. The two layers are assumed to be weakly interacting and do not affect each others equilibrium properties. The exchange-splitting field H_{ex} in the S layer acts on the spins of the electrons and breaks the particle-hole symmetry. The origin of this field can be due to the magnetic proximity effect with either a ferromagnetic insulator [25–29] [see Fig. 1(a)], or it can result from a Zeeman-field due to an applied magnetic field ($\vec{B}$) [17,30] [as shown in Fig. 1(b)]. In the linear response regime, one can write phenomenological TE coefficients in terms of the linear response matrices. For the total charge I_C and energy I_E currents

$$\begin{pmatrix} I_C \\ I_E \end{pmatrix} = \begin{pmatrix} G & P\alpha \\ P\alpha & G_{th}T \end{pmatrix} \begin{pmatrix} V \\ \Delta T/T \end{pmatrix}, \tag{1}$$

and for the total spin I_S and spin-energy I_{E_S} currents flowing through the tunnel contact

$$\begin{pmatrix} I_S \\ I_{E_S} \end{pmatrix} = \begin{pmatrix} PG & \alpha \\ \alpha & PG_{th}T \end{pmatrix} \begin{pmatrix} V \\ \Delta T/T \end{pmatrix}, \tag{2}$$

where G, α, P, G_{th}, and T are the charge conductance, TE coefficient, spin-polarization of the interface conductance, energy conductance, and the absolute temperature of the system, respectively. One can write down the linear-response matrices in terms of TE coefficients

$$G = G_T \int_{-\infty}^{\infty} dE N_0(E)\delta_T(E)\,, \tag{3a}$$

$$G_{th} = \frac{G_T}{e^2T} \int_{-\infty}^{\infty} dE N_0(E)E^2\delta_T(E)\,, \tag{3b}$$

$$\alpha = \frac{G_T}{2e} \int_{-\infty}^{\infty} dE N_z(E)E\delta_T(E)\,. \tag{3c}$$

Here, G_T is the conductance of the junction, $\delta_T(E) = [4k_BT\cosh^2(E/2k_BT)]^{-1}$, being the difference of the distributions functions of the F and S expanded with respect to a small temperature gradient ΔT across the junction, E is the quasiparticle energy and k_B is the Boltzmann constant. Total $N_0(E)$ and spin-polarized $N_z(E)$ DOS in S are defined as

$$N_0(E) = (N_\uparrow(E) + N_\downarrow(E))/2\,,$$
$$N_z(E) = N_\uparrow(E) - N_\downarrow(E)\,. \tag{4}$$

Based on Eqs. (3), the linear-response matrices depend on $N_0(E)$ and $N_z(E)$. These are evaluated employing the quasiclassical Green's function method. The charge Seebeck coefficient describes the buildup of a TE voltage caused by a temperature gradient throughout a material as induced by the Seebeck effect when one opens the circuit and the net electric current vanishes such that $I_C = 0$ [5]. From the knowledge of the electrical conductivity G and the TE coefficient α, one can define the charge Seebeck coefficient as $S = -P\alpha/(GT)$. Thus, an increase of the TE coefficient and a reduction of the charge conductance are required to enhance the thermopower. It is crucial to note that, a non-zero spin-polarization of the interface is necessary to observe the TE phenomena in the present system. According to Eqs. (3), we see that the charge

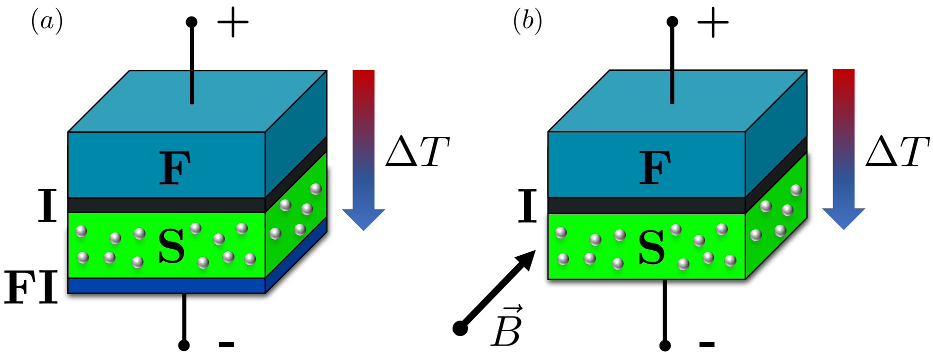

Figure 1. (Color online). Schematic illustration of the investigated thin-film heterostructure. The structure consists of a ferromagnet (top region in light-blue) coupled to a superconductor (bottom region in green) with the superconducting pair potential Δ through a thin insulating barrier (black region in the center). The quasiparticle DOS in the superconductor is modified by the presence of a spin splitting field H_{ex} which may be realized either (a) via proximity with a ferromagnetic insulator (FI) film (dark-blue region), or (b) an applied external magnetic field ($\vec{B}$). ∇T denotes the temperature gradient through the junction, from hot-end (F) to the cold-end (S). Presence of magnetic impurities inside the S results in spin-flip scattering.

conductance is proportional to the averaged of the spin tunneling DOS. However, the TE coefficient is a function of the difference between the Zeeman-split spin states $N_z(E)$. The spin-dependent thermopower [31–36] will be determined by the same parameters as the charge Seebeck coefficient and refers to the creation of spin voltage due to a temperature difference. In other words, spin thermopower is a way to quantify thermally produced spin voltages [37]. Eilenberger's equation of motion [23] for a spin-split superconductor is given by

$$[i(E \pm i\delta)\hat{\tau}_3 - \hat{\Delta} - iH_{ex}\sigma\hat{\tau}_3 - \hat{\Sigma}_{sf}, \hat{g}^{R/A}] = 0\,, \tag{5}$$

where we set $k_B = \hbar = 1$. $\hat{g}^{R/A}$ is the retarded (advanced) Green's function and can be obtained by replacing quasiparticle energy E by $[E \pm i\delta]$, respectively. Incoherent broadening δ, or the so-called 'Dynes' parameter [38] is modeling inelastic processes and has been taken into account in order to preserve the analytic structure of the Green's function [39]. Here, $\hat{\Delta}$ is the off-diagonal energy gap matrix given as $\hat{\Delta} = \Delta\hat{\tau}_1$ with $\hat{\tau}_i$ being the 2×2 Pauli matrix in Nambu space, and Δ is the superconducting pair potential which for simplicity is chosen to be real in our subsequent analysis. The superconducting pair potential at zero temperature, exchange field, and spin-flip scattering rate is denoted as Δ_0. The exchange field H_{ex} acts on the electron spins and splits the DOS for spin-up and spin-down electrons [40–42]. $\sigma = \{\pm 1\}$ is a spin label refering to the parallel and antiparallel orientations of the spin ($\uparrow / \downarrow$) with respect to the external spin splitting field. Since we have a homogeneous superconductor, the right-hand side of the equation (5) is equal to zero. The selfenergy corresponding to the spin-flip scattering $[\hat{\Sigma}_{sf} = (1/2\tau_{sf})\hat{\tau}_3\hat{g}^{R/A}\hat{\tau}_3]$ [42–46] accounts for spin-flip processes due to the presence of magnetic impurities inside the superconductor. Alternatively, such a selfenergy can originate from a magnetic interface with strong spin-dependent scattering [47–49] influencing the density of states [51, 52]. The spin-flip scattering time τ_{sf} is the average time between changes of the spin state of an electron [43]. We employ the following ϑ parametrization

$$\hat{g}^R = \cos\vartheta_\pm\hat{\tau}_3 + \sin\vartheta_\pm\hat{\tau}_1 = \begin{pmatrix} \cos\vartheta_\pm & \sin\vartheta_\pm \\ \sin\vartheta_\pm & -\cos\vartheta_\pm \end{pmatrix}, \tag{6}$$

which automatically ensures the normalization $\hat{g}^R\hat{g}^R = \hat{1}$. Here $\mathcal{G} = \cos\vartheta_\pm, \mathcal{F} = \sin\vartheta_\pm$, where $\mathcal{G}$ and $\mathcal{F}$ are normal and anomalous part of Green's functioms, and $\vartheta_\pm$ is a complex quantity.

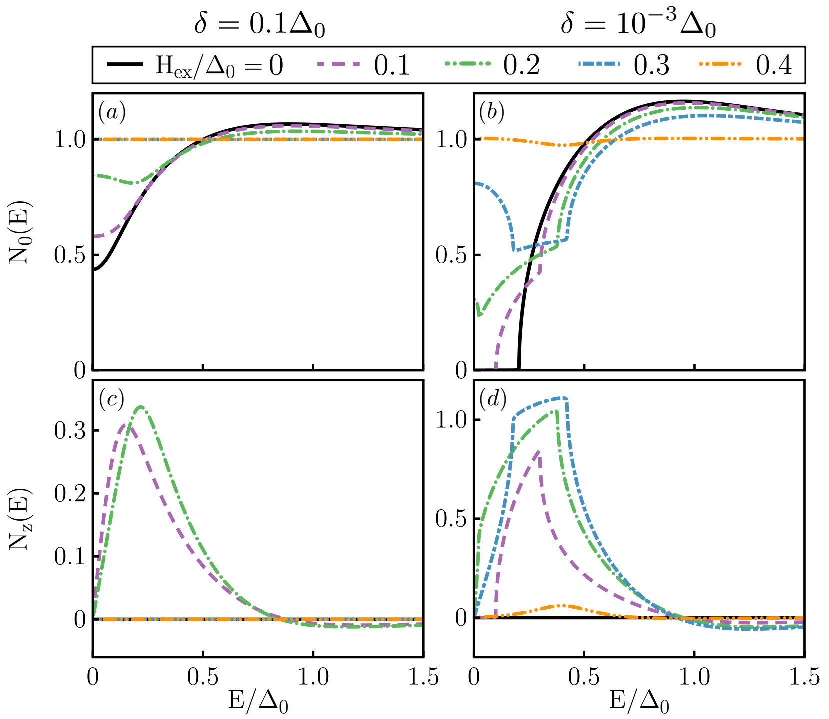

Figure 2. (Color online). Self-consistently determined quasiparticle DOS averaged over spin states $N_0(E)$ (Upper row) and their difference $N_z(E)$ (Lower row) with energies normalized to the superconducting pair potential at $T = 0, H_{ex} = 0$, and $\Gamma_{sf} = 0$ (Δ_0). The inverse spin-flip scattering rate is $\Gamma_{sf} = 0.3\Delta_0$ and temperature $k_BT/\Delta_0 = 0.1$ for $H_{ex}/\Delta_0 = 0$ (black solid line), 0.1 (purple dashed line), 0.2 (green dash-dot line), 0.3 (blue long dash-dash), 0.4 (orange dash-dot-dot). The left (right) panel is obtained for an incoherent broadening parameter $\delta = 0.1\Delta_0$ ($\delta = 10^{-3}\Delta_0$).

Using equations (5) and (6), one can simplify the equation of motion to a set of nonlinear equations for up and down-spins as

$$-2\big((E + i\delta) \pm H_{ex}\big) \sin\vartheta_\pm + 2i\Delta\cos\vartheta_\pm - i\Gamma_{sf}\sin 2\vartheta_\pm = 0\,, \tag{7}$$

with $\Gamma_{sf} = 1/\tau_{sf}$ being the spin-flip scattering rate. The superconducting pair potential is determined self-consistently from the imaginary component of the anomalous part (pair amplitude) of the retarded Green's function:

$$\Delta = \frac{\lambda}{2}\int_0^{\Omega_{\rm BCS}} dE \tanh\frac{E}{2k_BT}\Im[\sin\vartheta_+ + \sin\vartheta_-]\,, \tag{8}$$

where λ is the dimensionless Bardeen-Cooper-Schrieffer (BCS) [50] interaction constant, and for a weak interaction $\lambda \ll 1$. Once we have an expression for the self-consistent superconducting pair potential, we can calculate DOS from the real part of the normal component of the retarded Green's function. Within this framework, the DOS for spins up ($\uparrow$) and down ($\downarrow$) are

$$N_\uparrow(E) = N(0)\Re[\cos\vartheta_+]\,,$$
$$N_\downarrow(E) = N(0)\Re[\cos\vartheta_-]\,. \tag{9}$$

$N(0)$ is the DOS in the normal state at the Fermi level.

3. Results and discussions

Using equations (4) and (9), we calculate the DOS in S for averaged over spin $N_0(E)$ [see Fig. 2 (a)-(b)] and the difference of up and down-spin states $N_z(E)$ [see Fig. 2 (c)-(d)]. $N_0(E)$ and

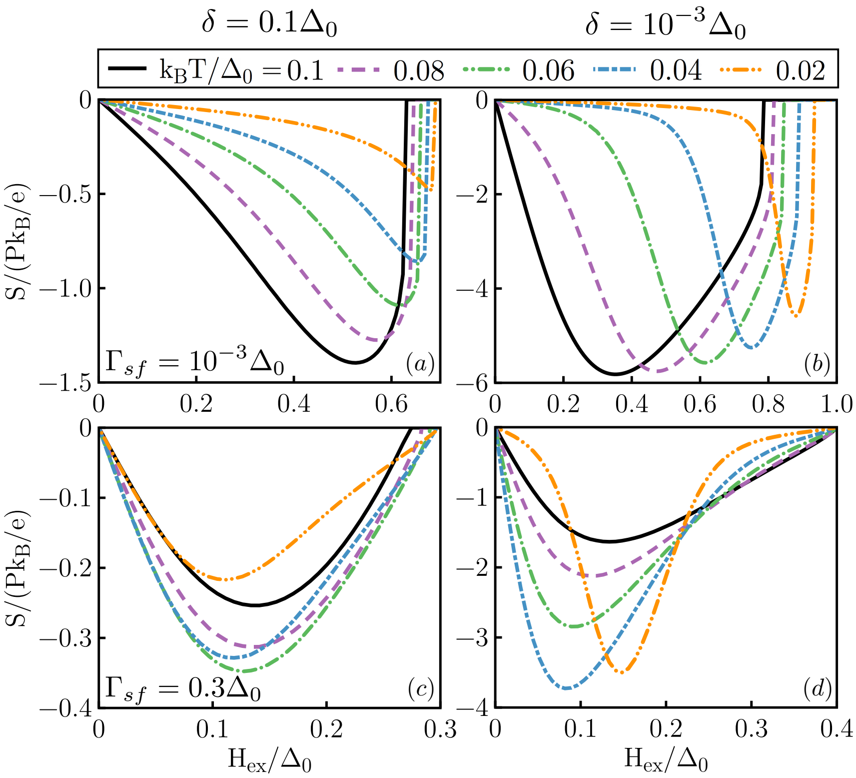

Figure 3. (Color online). Self-consistently determined charge Seebeck coefficient S normalized to Pk_B/e as a function of thew dimensionless spin splitting field H_{ex}/Δ_0. The curves are shown for different temperatures $k_BT/\Delta_0 = 0.1$ (black solid line), 0.08 (purple dashed line), 0.06 (green dash-dot line), 0.04 (blue long dash-dash) and 0.02 (orange dash-dot-dot). (a) and (b) in the top row correspond to $\Gamma_{sf} = 10^{-3}\Delta_0$, while for (c) and (d) in the bottom row the spin-flip rate is $0.3\Delta_0$. In the left panels we have set $\delta = 0.1\Delta_0$, while in the right panels the incoherent broadening is $10^{-3}\Delta_0$. The Seebeck coefficient increases with increasing the magnetic field, approaches a maximum for a specific field, and eventually drops to zero when the exchange field has suppressed completely the superconductivity and there is a phase transition to the normal state. Δ_0 is the zero-temperature, zero-exchange field, and zero-spin-flip scattering superconducting gap.

$N_z(E)$ are plotted in Fig. 2 for various values of spin splitting field at $k_BT/\Delta_0 = 0.1$ (here, Δ_0 is the superconducting pair potential at $T = 0, H_{ex} = 0$, and $\Gamma_{sf} = 0$). Figs. 2-(a) and 2-(c) demonstrate $N_0(E)$ and $N_z(E)$ evolution plotted for broadening parameter $\delta = 0.1\Delta_0$. Likewise, Figs. 2-(b) and 2-(d) show the DOS for $\delta = 10^{-3}\Delta_0$. On increasing the spin-flip scattering rate Γ_{sf}, the maximum of the tunneling DOS is shifted towards $E \ll \Delta_0$ thereby making interesting quasiparticle spin properties accessible in a lower energy range.

In Fig. 3, we plot the Seebeck coefficient $S/(Pk_B/e)$ vs. the spin splitting field H_{ex}/Δ_0 for different temperatures. Figs. 3-(a) and 3-(b) compare the Seebeck coefficient at $\Gamma_{sf} = 10^{-3}\Delta_0$ for two values of incoherent broadening $\delta = 0.1\Delta_0$ and $10^{-3}\Delta_0$. Correspondingly, Figs. 3-(c) and 3-(d) show the thermopower obtained for $\Gamma_{sf} = 0.3\Delta_0$. The qualitative behavior is analogous to the previous case, but the main notable difference is that the spin-flip processes strongly enhance the thermopower maximum and consequently the TE performance of the system in the low-field and low-temperature regime.

Fig. 4 shows the charge Seebeck coefficient $S/(Pk_B/e)$ as a function of inverse scattering rate Γ_{sf} at $H_{ex}/\Delta_0 = 0.2$ for different temperatures calculated for incoherent broadening $\delta = 0.1\Delta_0$ [see Fig. 4-(a)], and $\delta = 10^{-3}\Delta_0$ [see Fig. 4-(b)]. It is interesting to note that by increasing the incoherent broadening, we can access a larger thermopower in the low-temperature regime.

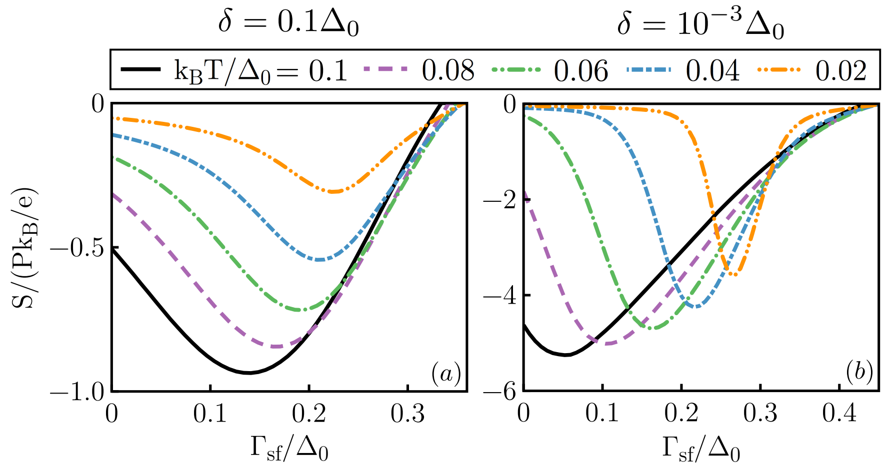

Figure 4. (Color online). Self-consistently determined plot of the normalized thermopower $S/(Pk_B/e)$ versus spin-flip rate Γ_{sf} calculated for $H_{ex}/\Delta_0 = 0.2$ at same selected temperatures as in Fig. 3, $k_BT/\Delta_0 = 0.1$ (black solid line), 0.08 (purple dashed line), 0.06 (green dash-dot line), 0.04 (blue long dash-dash) and 0.02 (orange dash-dot-dot). The curves have been calculated for (a) the finite incoherent broadening $\delta = 0.1\Delta_0$ and (b) $\delta = 10^{-3}\Delta_0$. Δ_0 is the superconducting pair potential at zero temperature, zero exchange field, and zero spin-flip scattering rate.

In order to illustrate the effect of the incoherent broadening on the TE performance of our tunnel contact, in Fig. 5 we present the result of the maximal thermopower $|S/(Pk_B/e)|_{max}$ calculated by varying the magnetic field at $k_BT/\Delta_0 = 0.02$ for different broadening values. It can be seen that the curve calculated for $\delta = 0.1\Delta_0$ drastically reduces as a function of spin-flip scattering rate Γ_{sf} compared to the one which is calculated for $\delta = 10^{-3}\Delta_0$. Another interesting effect that one observes is that $|S/(Pk_B/e)|_{max}$ calculated for $\delta = 10^{-3}\Delta_0$ exceeds k_B/e by the factor of 4.6, which is approximately about an order of magnitude larger than the value calculated for a larger incoherent broadening parameter. It is interesting to note that the spin-flip rate does not effect significantly the maximum of thermopower for $\delta = 10^{-3}\Delta_0$, however, it shifts the maximal value from a high field $H_{ex}/\Delta_0 \approx 0.9$ to a lower field $H_{ex}/\Delta_0 \approx 0.15$.

4. Conclusions

In conclusion, we have investigated the thermoelectric response of a tunnel structure formed by a thin insulating layer sandwiched between a spin-split superconducting film and a ferromagnetic metal. We take into account the important role of spin-flip scattering and incoherent broadening. All calculations have been done self-consistently with quasiclassical Green's function technique. We have shown that increasing the spin-flip scattering rate leads to strong enhancement of the TE performance in the low-field and low-temperature regime. Specifically, the maximum of thermopower exceeds k_B/e by a factor of 4.6 for $k_BT/\Delta_0 = 0.02$, and has a nonmonotonic behaviour with respect to the spin splitting and the spin-flip scattering rate in the superconductor. We further have found that the incoherent broadening leads to a drastic reduction of the TE performance [see Fig. 5]. Although spin-flip rate does not affect the maximum of the thermopower, it shifts the peak from a higher to a lower field. Our results constitute a promising step towards reducing the necessary spin splitting in these structures.

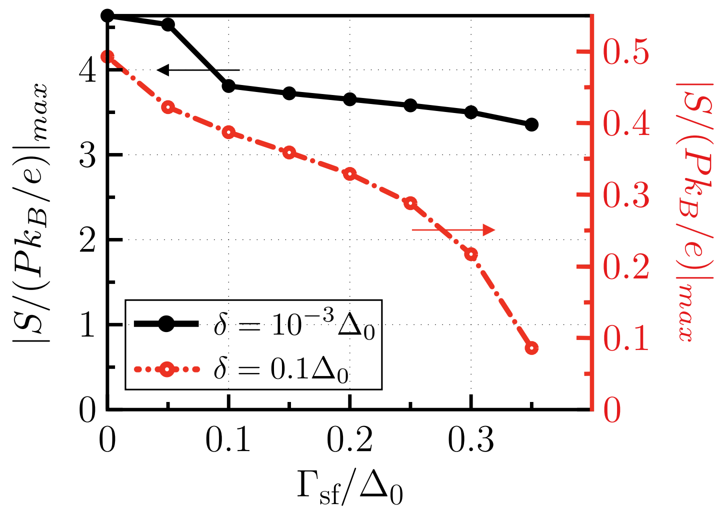

Figure 5. (Color online). Comparison between the maximal thermopower $|S/(Pk_B/e)|_{max}$ vs spin-flip rate Γ_{sf} calculated by varying the magnetic field at $k_BT/\Delta_0 = 0.02$ [see the orange curve in Fig. 3]. The solid and dash-dotted line have been computed for the incoherent broadening $\delta = 10^{-3}\Delta_0$ and $0.1\Delta_0$, respectively. Colored arrows indicate the corresponding thermopower axis to each δ. Δ_0 is the energy gap at $T = 0, H_{ex} = 0$, and $\Gamma_{sf} = 0$.

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