Spin-flip enhanced thermoelectricity in superconductor-ferromagnet bilayers

A Rezaei, A Kamra, P Machon and W Belzig
Department of Physics, University of Konstanz, D-78457 Konstanz, Germany
E-mail: ali.rezaei@uni-konstanz.de

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Abstract
We study the effects of spin-splitting and spin-flip scattering in a superconductor (S) on the thermoelectric (TE) 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 is attributed to the spin-dependent particle–hole asymmetry in the tunneling density of states (DOS) in the S/F heterostructure. Here, we evaluate the S DOS and thermopower for a range of temperatures, Zeeman-splitting, and spin-flip scattering. In contrast to the naive expectation based on the negative effect of spin-flip scattering on Cooper pairing, we find that the spin-flip scattering strongly enhances the TE 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 $\approx 5$ and has a non-monotonic dependence on spin-splitting and spin-flip rate.

1. Introduction

The field of thermoelectrics (TEs) 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 TE or Seebeck effect refers to the conversion of a temperature difference ($\Delta T$) across a system into the 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 the density of states (DOS) variations versus quasiparticle energy giving rise to a net charge current. Under open circuit conditions, the requirement of no net current leads to a charge buildup that appears as a voltage called thermopower. However, the variation in the DOS is typically very small and, consequently, the TE 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 spin-splitting breaks the spin-dependent electron-hole symmetry. Its interface with a ferromagnetic metal results in a spin-polarized conductance leading to a large Seebeck effect. Following this principle, large spin-dependent TE effects have been theoretically proposed [2–9] and experimentally demonstrated [10–12] in spin-split and -polarized superconducting tunnel contacts [13]. Furthermore, TE properties in a variety of systems have been investigated [14–16]. In most of the cases, the Zeeman-split superconducting heterostructures exhibiting sizeable TE effects require a large externally applied magnetic field [10, 17]. A recent experiment [11] reported a large Seebeck coefficient employing the spin-splitting field provided by the proximity to a ferromagnetic insulator, instead of an applied magnetic field. The spin–orbit interaction has been found to reduce the TE performance of such structures [18]. The usability of these devices as thermal detectors has also been discussed [19].

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Spin-splitting required to achieve a large Seebeck coefficient rapidly as the superconductivity is destroyed by an increasing spin-superconducting gap. However, the TE response is substantial even in the gapless regime and starts to diminish.

We consider a ferromagnetic metal

\[ \text{2. Model} \]

Figure 1. 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) 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 which may be realized via either (a) proximity with a ferromagnetic insulator (FI) film (dark-blue region), or (b) an applied external magnetic field. \( \Delta T \) denotes the temperature gradient through the junction. The presence of magnetic impurities inside the superconductor or scattering at its interface with the magnetic insulator results in spin-flip processes.

Similar to the influence of the spin–orbit interaction \[18\], spin-flip scattering may be expected to reduce the TE performance of such devices. This is because spin-flip scattering is known to have a detrimental effect on superconductivity via breaking of the Cooper pairs resulting in a reduction of the gap. One may assume a different perspective and argue that the reduction of the superconducting gap might enable quasiparticles at lower energies to contribute towards the Seebeck effect, thereby enhancing it. Thus, competing mechanisms are in play and a clear winner may not be ascertained \textit{a priori}. Earlier works have investigated the influence of spin-flip on the critical current \[20\] and critical temperature \[20, 21\], the spatial and energy dependences of the DOS \[22–25\], the anomalous Green’s function \[26\], and quasiparticles distribution \[27, 28\] in a spin-split S.

In this work, we theoretically address the effect of spin-flip scattering on the Seebeck coefficient in such S/F bilayers. We find that spin-flip enhances the TE response at low values of the spin-splitting. We incorporate spin-flip scattering and incoherent broadening self-consistently within the quasiclassical description of S \[25, 29\]. The spin-flip scattering shifts the peaks of the DOS toward zero energy (see figure 2) thus reducing the gap and eventually leading to gapless superconductivity. We find that spin-flip scattering suppresses (enhances) the Seebeck coefficient at-large (low) spin-splitting of the DOS. We find that the maximal Seebeck coefficient is relatively insensitive to the spin-flip rate, while the value of spin-splitting at which this maximum is achieved is reduced by increasing the spin-flip rate. Thus, for weak spin-flip scattering, its major effect is to decrease the spin-splitting required to achieve a large Seebeck coefficient, corresponding to the reduction of the superconducting gap. However, the TE response is substantial even in the gapless regime and starts to diminish rapidly as the superconductivity is destroyed by an increasing spin-flip rate. We find an analogous behavior of the figure of merit \( ZT \).

This paper is organized as follows. In section 2, we outline the model and theoretical framework employed to describe the TE effects in an S/F bilayer. We present and discuss our results regarding the dependence of the Seebeck coefficient on the spin-flip scattering rate in section 3. We conclude by summarizing our findings in section 4.

\[ \text{2. Model} \]

We consider a ferromagnetic metal (F) coupled via a spin-polarized tunneling contact to a spin-split superconductor (S) layer. A sketch of the device under consideration has been shown in figure 1. The spin-splitting in S may be caused by proximity effect with a ferromagnetic insulator \[30–34\] (figure 1(a)) or an applied magnetic field \( \vec{B} \) \[17, 35\] (figure 1(b)). The spin-splitting field is oriented collinear to the magnetization direction of F. The thickness of the superconducting film is assumed smaller than the superconducting coherence length. The coupling between the two layers is assumed to be weak enough not to affect each others equilibrium properties. The exchange-splitting field \( H_{\text{ex}} \) in the S layer acts on the spins of the electrons and breaks the particle–hole symmetry in the spin-resolved DOS. Considering the drives to be small, one can write TE coefficients in terms of the linear-response matrices.

Adopting the notation of \[5\], we have for the charge \( I_c \) and energy \( I_E \) currents

\[
\begin{pmatrix}
    I_c \\
    I_E
\end{pmatrix} =
\begin{pmatrix}
    G & P \alpha \\
    P \alpha & G_{\theta \theta}
\end{pmatrix}
\begin{pmatrix}
    V \\
    \Delta T/T
\end{pmatrix},
\]

\[ \text{1} \]
and for spin $I_2$ and ‘spin-energy’ $I_{E_2}$ currents flowing through the tunnel contact

$$
\begin{align*}
    \left( \frac{I_S}{I_{E_2}} \right) &= \left( \frac{PG}{\alpha} \right) \frac{\epsilon}{PG_{th} T} \left( \frac{V}{\Delta T / T} \right).
\end{align*}
$$

(2)

where $G$, $\alpha$, $P$, $G_{th}$, and $T$ are the charge conductance, TE coefficient, spin-polarization of the tunnel contact, thermal conductance, and the absolute temperature of the system, respectively. The different coefficients may further be expressed in terms of the spin-resolved DOS:[5]

$$
G = G_T \int_{-\infty}^{\infty} dE N_0(\epsilon) \delta_T(\epsilon),
$$

(3a)

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

(3b)

$$
\alpha = \frac{G_T}{2e} \int_{-\infty}^{\infty} dE N_0(\epsilon) E \delta_T(\epsilon).
$$

(3c)

Here, $G_T$ is the conductance of the junction in the normal state, $E$ is the quasiparticle energy, $k_b$ is the Boltzmann constant, and $\delta_T(\epsilon) \equiv [4k_b T \cosh^2 (E/2k_b T)]^{-1}$ being the normalized difference of the quasiparticle distributions functions of the F and S expanded with respect to a small temperature difference $\Delta T$ across the junction. The total and the spin-polarized DOS in $S N_0(\epsilon)$ and $N_\sigma(\epsilon)$, respectively, are defined as

$$
N_\sigma(\epsilon) = (N_\uparrow(\epsilon) + N_\downarrow(\epsilon))/2,
$$

(4)

$$
N_\sigma(\epsilon) = N_\uparrow(\epsilon) - N_\downarrow(\epsilon),
$$

where $N_{\uparrow(\downarrow)}$ is the DOS for spin-up (down) quasiparticles in S. These are evaluated employing the quasiclassical Green’s function method as described below.

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] Employing equation (3a), the charge Seebeck coefficient is obtained 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. From equation (3a), we see that the charge conductance is proportional to the averaged tunneling DOS $N_\sigma(\epsilon)$. However, the TE coefficient is a function of the spin-polarized DOS $N_\sigma(\epsilon)$. In the system under investigation, the spin Seebeck effect [36–41] is 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, the spin thermopower is a way to quantify thermally produced spin voltages [42]. In the linear-response regime, the efficiency of the device to produce TE power is quantified by the so-called dimensionless figure of merit $ZT$: $ZT = S^2 GT / G_{th}$, where $G_{th} = G_{th} - (P\alpha)^2 / GT$ is the energy conductance at zero current [5]. The larger the value of the $ZT$ the more efficient is the TE device [43].

The Eilenberger equation of motion [29] for a homogeneous spin-split superconductor is given by

$$
[iE \delta - \hat{\Delta} - iH_{ex} \sigma \hat{\gamma} - \hat{\Sigma}_{sd}, \hat{g}_R^{R/A}] = 0,
$$

(5)

where we have set $k_b = \hbar = 1$. $\hat{g}_R^{R/A}$ is the retarded (advanced) Green’s function and is obtained by replacing the quasiparticle energy $E$ by $[E \pm i\delta]$, respectively. The incoherent broadening $\delta$ or the so-called ‘Dynes’ parameter [44], parameterizes inelastic scattering and conserves the analytic structure of the Green’s functions [25]. Here, $\hat{\Delta}$ is the superconducting pair potential matrix given as $\hat{\Delta} = \Delta \hat{\gamma}$, with $\gamma$ the Pauli matrices in Nambu space. For a homogeneous superconducting state, $\Delta$ may be chosen as real and corresponds to the superconducting gap. The gap at zero temperature, exchange field, and spin–flip scattering rate is denoted by $\Delta_0$. The exchange field $H_{ex}$ acts on the electron spins and splits the DOS for spin-up and spin-down electrons [45–47]. $\sigma = \{\pm\}$ is a spin label referring to the up and down orientations of the spin (\(\uparrow/\downarrow\)) with respect to the external spin–splitting field. Since we neglect inhomogeneities, gradient–terms do not appear in equation (5). The self-energy corresponding to spin–flip scattering via magnetic impurities [47–51] corresponds to $\hat{\Sigma}_{sd} = (1/2\tau_\sigma) \hat{\gamma} \hat{g}_R^{R/A} \hat{g}_R^{R/A}$. Alternatively, such a self-energy can originate from a magnetic interface with strong spin-dependent scattering [52–54] thereby influencing the DOS [55, 56]. The spin–flip scattering time $\tau_\sigma$ is the average time between changes of the spin state of an electron [48].

We employ the following $\theta$ parameterization for the quasiclassical Green’s function matrix

$$
\hat{g}_R^{R,A} = \cos \vartheta^R \hat{g}_R^{R} + \sin \vartheta^R \hat{g}_L^{R},
$$

(6)

which automatically ensures the normalization $\hat{g}_R^{R,A} \hat{g}_R^{R,A} = \hat{1}$. In these expressions $\cos \vartheta^R$ and $\sin \vartheta^R$ are normal and anomalous Green’s functions, respectively, and $\vartheta^R$ is, in general, a complex quantity. Using equations (5) and (6), the equation of motion simplifies to a set of nonlinear equations for up and down-spins as
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:

$$\Delta = \frac{\lambda}{2} \int_{0}^{\Omega_c} dE \tanh \frac{E}{2k_B T} \mathcal{J} [\sin \vartheta_+ + \sin \vartheta_-],$$

where $\lambda$ is the dimensionless Bardeen–Cooper–Schrieffer [57] interaction constant. $\Omega_c$ is a suitably chosen cut-off. Once we have an expression for the self-consistent superconducting pair potential, we can calculate the DOS from the real part of the normal component of the retarded Green’s function. Within this framework, the DOS for spin-up ($\uparrow$) and down ($\downarrow$) are

$$N_{\uparrow}(E) = N(0) \Re \{\cos \vartheta_+\},$$
$$N_{\downarrow}(E) = N(0) \Re \{\cos \vartheta_-\},$$

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

Our approach outlined above determines the relevant properties of the superconducting state via the self-consistency equation, and then employs the knowledge of these equilibrium properties to evaluate the system response. However, this approach does not guarantee that the equilibrium state thus obtained is lowest in energy, and hence, the ‘true’ ground state. For a superconductor in a magnetic field, the superconducting ground state, disregarding spin-flip scattering, is stable, i.e. lowest in energy, only below the so-called ‘Chandrasekhar–Clogston’ (CC) limit $H_{ex} = \Delta_0/\sqrt{2}$ [58], which is lower than the Pauli limit $H_{ex} = \Delta_0$ captured by our approach. Hence, our calculations pertain to the superconducting solution of the self-consistency equation, which becomes metastable in certain range of exchange-splitting, i.e. its energy is larger than the system being in normal state. The true ground state in this range is the normal state, the TE response in which is vanishingly small. We determine this range from the literature [18, 59] and mark it via shaded regions in figures 3, 4 and 6.

### 3. Results and discussions

In figure 2, we show the total [$N_0(E)$] and spin-polarized [$N_{\uparrow}(E)$] DOS obtained within a self-consistent evaluation for different exchange fields and a finite spin-flip scattering rate as described above. For the total DOS, the gap slowly closes as the spin-splitting is increased, and via the gapless state, eventually results in the destruction of the superconducting state. The spin-polarized DOS vanishes for no spin-splitting as well as at-large spin-stittings, when the superconducting state is destroyed. Hence, we see that the optimal situation for the TE coefficient $\alpha$, corresponding to the largest $N_{\uparrow}(E)$ (equation (3a)), is achieved for a certain value of the spin-splitting.

Employing the obtained DOS and equations (3a), we evaluate the Seebeck coefficient and plot it versus spin-splitting in figure 3. As expected from our earlier discussion on spin-polarized DOS, the Seebeck coefficient vanishes at zero and large spin-splittings yielding a maximum in between. We see that the value of spin-splitting at which the maximal Seebeck coefficient is achieved varies strongly with the temperature. Remarkably, in the
absence of spin-flip scattering (left panel of figure 3) at the lowest temperatures, the Seebeck coefficient is drastically reduced due to the CC restriction on the field range.

Now comparing the cases of small and large spin-flip rate in figure 3, we observe that a large spin-flip rate reduces the temperature sensitivity of the spin-splitting ($H_{\text{max}}$) corresponding to a maximal Seebeck effect. Overall, putting the CC physics aside for the moment, $H_{\text{max}}$ lowers with an increasing spin-flip rate, but this lowering does not have a one-to-one correspondence with the reduction of the superconducting gap, as evidenced by the relative insensitivity of $H_{\text{max}}$ to the temperature. The TE performance of the device is particularly insensitive to the temperature for large spin-flip, as long as the system is not in the gapless state.

Figure 4 presents the analogous results for $ZT$ as a function of the normalized spin-splitting field $H_{\text{ex}}/\Delta_0$. The parameter values employed are $\Gamma_s/\Delta_0 = 10^{-3}$ and 0.3 for left and right panels, respectively. We have set the incoherent broadening $\delta = 10^{-3}\Delta_0$. 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 destroyed the superconductivity. The shaded region depicts the exchange fields above the CC limit. Thus, the plotted results correspond to the metastable superconducting state, while the true stable state is normal and corresponds to vanishing thermoelectric response.

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Figure 4 presents the analogous results for $ZT$ corroborating the similar dependence of TE efficiency on the relevant physical variables. We emphasize that $ZT$ values larger than 1 can only be achieved in the low-temperature and low-exchange field regime by introducing a spin-flip rate as shown in the right panel of figure 4. In the absence of spin-flip scattering, $ZT$ is dramatically reduced at very low temperatures $T \lesssim 0.05\Delta_0$ due to the instability of the superconducting state in the CC regime.

We now focus on the dependence of the TE response on the spin-flip rate. To this end, we plot the Seebeck coefficient (left panel) and $ZT$ (right panel) versus spin-flip rate at a fixed value of spin-splitting in figure 5. The Seebeck coefficient exhibits an initial enhancement with the spin-flip rate followed by an eventual reduction. This behavior may be understood in terms of spin-flip mediated lowering of the superconducting gap. The first order effect of spin-flip in this regime is to reduce the superconducting gap thereby shifting the quasiparticle
dynamics to lower energies without significantly damaging the superconducting state. At larger spin-flip rates, the Cooper pair depairing effect dominates the modification of the system TE response. $ZT$ essentially mirrors the dependence of Seebeck coefficient with minor differences.

We finally study the dependence of the maximum Seebeck coefficient (as a function of spin-splitting) on the spin-flip rate in figure 6. The maximum Seebeck coefficient is relatively insensitive to the spin-flip rate. However, the value of spin-splitting (not shown explicitly) at which this maximum value is achieved decreases monotonically with the spin-flip rate.

Based on the analysis presented above, we are now in a position to summarize the effect of spin-flip on the TE response of the system under investigation. At small values of the spin-flip scattering rate, its predominant effect is reducing the superconducting gap. This provides quasiparticles at lower energies and results in enhancement of Seebeck effect at low values of the spin-splitting exchange field. After the superconductor enters the gapless regime, there is no such gain in further increasing the spin-flip. On the contrary, the depairing effect only tends to make the DOS flatter resulting in a decreasing TE response with increasing spin-flip. We note that spin–orbit scattering will mix the spin-split energy bands \[59\] and hence reduce the overall TE performance. Thus, the optimal value of spin-flip is around the point when the superconductor is about to enter the gapless regime.

Figure 5. Spin–flip scattering rate ($\Gamma_{sf}/\Delta_0$) dependence of the self-consistently determined normalized thermopower $S/(PkB/e)$ (left panel) and thermoelectric figure of merit $ZT$ (right panel) for $H_{ex}/\Delta_0 = 0.2$ at different temperatures. Polarization of the interface conductance and incoherent broadening are assumed $P = 0.9$ and $\delta = 10^{-3}\Delta_0$.

Figure 6. Maximal thermopower $|S/(PkB/e)|_{max}$ versus spin–flip rate $\Gamma_{sf}/k_BT/\Delta_0 = 0.02$ (the orange curves in figure 3). For a small incoherent broadening $\delta = 10^{-3}\Delta_0$, the maximum Seebeck coefficient for medium spin–flip scattering rates is relatively insensitive to the spin–flip rate. The shaded region represents the destruction of the superconducting state due to CC physics. The upper spin–flip rate limit of the shaded region has not been calculated exactly and represents an approximate value. At small spin–flip rates, the true Seebeck coefficient vanishes due to the required exchange field being above the CC limit. On the other hand, large spin–flip rates destroy superconductivity via the usual depairing channel.
4. Conclusions

We have investigated the TE 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 roles of the spin-flip scattering and CC paramagnetic limit [58, 59]. The relevant physical quantities have been obtained self-consistently within the quasiclassical Green’s function technique. We have shown that increasing the spin-flip scattering rate leads to a strong enhancement of the TE performance in the low-field and low-temperature regime. Specifically, the maximum of the thermopower exceeds $k_B/e$ by a factor of ≈5 down to temperatures of the order of $k_B T / \Delta_0 \approx 0.02$ and has a non-monotonic behavior with respect to the spin-splitting and the spin-flip scattering rate in the superconductor. We also predict a sizeable TE figure of merit $ZT > 1$ in the low-temperature regime. Although the spin-flip rate does affect the maximum of the thermopower only weakly, it shifts the peak from a higher to a lower field. This appears to be of crucial importance since we find that the high fields required in the absence of spin-flip scattering are larger than the CC limit, and thus do not support superconductivity. Our results constitute a promising prediction for reducing the necessary spin-splitting in these structures, which might also be useful to avoid other detrimental effects related to external magnetic fields, magnetization control or the CC-limited ultra-low-temperature degradation of the TE response.

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ORCID iDs

A Rezaei @ https://orcid.org/0000-0001-9150-9520
A Kamra @ https://orcid.org/0000-0003-0743-1076
W Belzig @ https://orcid.org/0000-0002-5109-2203

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