Modelling of the Peltier effect in magnetic multilayers

Isaac Juarez-Acosta,1 Miguel A. Olivares-Robles,2 Subrojati Bosu,3,4 Yuya Sakuraba,4 Takahide Kubota,3 Saburo Takahashi,3 Koki Takanashi,3 and Gerrit E. W. Bauer3,5,6

1SEPI ENCB, Instituto Politecnico Nacional, Mexico D.F. 11340, Mexico
2SEPI ESIME Culhuacan, Instituto Politecnico Nacional, Mexico D.F. 04430, Mexico
3Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
4National Institute for Materials Science, Ibaraki 305-0047, Japan
5WPI-AIMR, Tohoku University, Sendai 980-8577, Japan
6Kavli Institute of Nanoscience, Delft University of Technology, Lorentzweg 1, 2628 CJ Delft, The Netherlands

(Dated: June 10, 2015)

We model the charge, spin, and heat currents in ferromagnetic metal/normal metal/normal metal trilayer structures in the two current model, taking into account bulk and interface thermoelectric properties as well as Joule heating. Results include the temperature distribution as well as resistance-current curves that reproduce the observed shifted parabolic characteristics. Thin tunneling barriers can enhance the apparent Peltier cooling. The model agrees with experimental results for wide multilayer pillars, but the giant effects observed for diameters ≲ 100 nm are still under discussion.

I. INTRODUCTION

Thermoelectric effects result from the coupling between energy and particle transport in conductors. An example is the heat current that is associated with a charge current and proportional to a material parameters called Peltier constant. At a thermocouple, i.e. a junction between two conductors with different Peltier coefficients, the heat current is not conserved, which implies heating or cooling depending on the current direction. The thermopower, on the other hand, is the thermoelectric voltage that is generated by a temperature difference over a metal wire that is proportional to the Seebeck coefficient. A thermocouple generates an isothermal thermoelectric voltage proportional to the difference between the Seebeck coefficients when the end of the wires are maintained at a temperature different from the junction. Nanostructured materials can enhance the efficiency of thermoelectric devices. Thermoelectric effects in metallic heterostructures including ferromagnets depend on the spin degree of freedom. The spin dependence of thermoelectric cooling is part of the field that studies the coupling between spin, heat and electric transport in small structures and devices, or spin caloritronics. Heating is an important issue for spin torque magnetic random access memories (STT-MRAM) device [MRAM], and spin caloritronic effects can improve their performance.

An enhanced Peltier effect has been reported by Fukushima et al. in metallic multilayers when structured into nanopillars. The effect was detected by change $\Delta R$ of the resistance $R_0$ as a function of current bias that acted as a thermometer. The Peltier effect cools or heats the systems by a term linear to the applied charge current $I_c$ and Peltier coefficient $\Pi$, while the Joule heating induces a temperature and resistance change that scales like the square of the current bias such that $\Delta R \sim R_0 I_c^2 - \Pi I_c$. At small currents the linear term dominates and causes a reduction of the resistance, i.e. an effective cooling, that in some structures was found to be very large. The Peltier coefficient was found by measuring the current where heating and cooling compensate each other and $\Delta R \left( I_c^{(0)} \right) = 0$ and therefore $\Pi = R_0 I_c^{(0)}$. The observed $\Pi = 480$ mV in pillars containing Constantan is attractive for cooling nanoelectronic devices. The cooling power enhancement was tentatively explained by Yoshida et al. by adiabatic spin-entropy expansion. However, such an equilibrium cooling mechanism could not explain that $\Pi$ is material dependent and even changes sign. The diffusion equation approach by Hatami et al. did take not into account either the precise sample configuration nor Joule heating and could not reproduce the large observed effects. The physical mechanism of the giant Peltier effect therefore remains unexplained. On the other hand, the recent experiments by Bosu et al. confirmed large Peltier coefficients for pillars including Heusler alloys when becoming very narrow. The present research has been motivated by the wish to model the heat and charge currents realistically in the hope to shed light onto this quandary. We report detailed calculations for the structure and model parameters matching Bosu et al. experiments and compare results of semi-analytic calculations with experiments. This study is limited to thermoelectric effects as described by the two-current model of thermoelectric transport in which spin current is carried by particle currents. We do not include explicitly phonon contributions to the heat current as well as phonon/magnon drag effects on the thermoelectric coefficients, which may lead to a temperature dependence of the model parameters. Furthermore, we completely disregarding collective effects that give rise to e.g. the spin Seebeck and spin Peltier effects. There are no indications that these approximations will do more than leading to some renormalization of the model parameters. While we are still far off a complete understanding of the experiments, we find evidence that very thin (Ohmic) tunnel junctions can enhance the Peltier effect.

This paper is organized as follows. In Section II, we re-
view the standard Valet-Fert model for spin transport in our nanopillars, with explicit to inclusion of interfaces. In Section III, we extend the model to include heat currents, charge and spin Joule heating, and explain our method to compute temperature profiles. In Section IV, we present results for the Peltier effect due different interfacial thermoelectric parameters and simulations of the Peltier effect are also performed, illustrating the importance of interface resistances, to finish in section V with a summary and conclusions.

II. SPIN-DEPENDENT DIFFUSION IN F|N|N_B MODEL

Our model can be applied quite generally to arbitrary multilayered structures, but we focus here on the charge-current biased trilayer nanostructures measured by Bosu et al. The thicknesses of F, N and N_B are L_F, L and L_B, respectively, and the device is sandwiched between two thermal reservoirs at same temperature T_0. The electric, spin and heat transport is described by an extended Valet-Fert model, including interfaces and spin-dependent thermoelectric effects. The parameters are interfaces resistances R_i and R_2 for interfaces the F[N and N|N_B respectively, bulk resistance R_i (i = F, N, N_B) for each metal, as well as the spin polarization P_F of the ferromagnetic metal.

We adopt a one-dimensional diffusion model in which the current flows along the x-direction and the origin is at the F|N interface. In the collinear two-channel resistor model, the electrons are in either spin-up or spin-down states. We divide the structure into various elements such as resistors, nodes and reservoirs. Discrete resistive elements are interfaces, tunnel barriers or constrictions that limit the transport. For our purpose, resistors are separated by nodes in which electrons can be described semiclassically by distribution functions \( f_i \). If the interactions electron-electron or electron-phonon are sufficiently strong, \( f_i \) approaches the Fermi-Dirac distribution which depends on temperatures T_i and chemical potentials \( \mu_i \). We disregard spin-dependent temperatures here but allow for spin accumulations, i.e. local differences between chemical potentials for both spins.

The spin particle \( I^{(a)}_c \) and heat \( J^{(a)}_q \) currents at a position \( x \) in a resistive element are

\[ I^{(a)}_c = A_c \int d\epsilon j^{(a)}(\epsilon, x) \] (1)

and

\[ J^{(a)}_q = -\frac{1}{e} \int d\epsilon j^{(a)}(\epsilon, x) - \mu_0 \int d\epsilon j^{(a)}(\epsilon, x) \] (2)

respectively, where \( A_c \) is the cross sectional area of the nanopillar, \( \alpha = \uparrow, \downarrow \) is the electron spin degree of freedom, \( j^{(a)} \) is the spin, energy (\( \epsilon \)) and position (\( x \)) dependent spectral current density, and \( \mu_0 \) the ground-state chemical potential. \( j^{(a)} = \sigma^{(a)}(\epsilon) f^{(a)}(\epsilon, x) \) is described by local Fermi-Fert distributions \( f^{(a)} \) at temperature T and spin-dependent chemical potentials \( \mu^a \), times the energy-dependent conductivity \( \sigma^a(\epsilon) \).

The spin accumulation is defined as \( \mu_s = \mu_\uparrow - \mu_\downarrow \), where \( \mu_\uparrow, \downarrow \) are the spin-up and spin-down chemical potential of the material, while the charge chemical potential is the average of the sum of spin-up and spin-down chemical potentials \( \mu_c = (\mu_\uparrow + \mu_\downarrow)/2 \).

The transport in each layer is governed by spin and charge diffusion equations, given by

\[ \frac{\partial^2}{\partial x^2} \mu_s = \frac{\mu_s}{\lambda^2} \] (3)

\[ \frac{\partial^2}{\partial x^2} \mu_c = -P_F \frac{\mu_s}{2\lambda^2} \] (4)

where \( \lambda \) is the spin-flip diffusion length, usually much larger in normal metals than in ferromagnetic metals \( \lambda_{N,N_B} \gg \lambda_F \), and

\[ P_F = \frac{\sigma_\uparrow - \sigma_\downarrow}{\sigma_\uparrow + \sigma_\downarrow} \] (5)

which is the spin polarization of conductivity in terms of the spin-dependent conductivity for each channel in the ferromagnet. In normal metals N and N_B these polarizations vanish (\( P_{N,N_B} = 0 \)). The solution of Eq. (3) depends on the integration constants Y and Z. For a ferromagnetic metal F, we find (see Fig. 1)

\[ \frac{\mu_c^{(a)}(x)}{e} = -\frac{I_c}{\sigma_F A_c} x + \alpha_1 + \frac{\sigma_F}{\sigma_F^c} \beta_1 e^{\frac{x}{\lambda_F}} \] (7)

where \( \alpha_1 \) is the voltage drop at the interface F|N, \( \sigma_F \) is the electrical conductivity of the ferromagnetic metal,

![Diagram](image-url)
For the normal metals, the spin-up and spin-down chemical potentials read

$$\mu_N^x(x) = -\frac{1}{\sigma_N A_c} x + b_1 e^{-\frac{x}{\lambda_N}} \pm b_2 e^{\frac{x}{\lambda_N}} \tag{8}$$

$$\mu_{N B}^x(x) = -\frac{1}{\sigma_{N B} A_c} (x - L) + \alpha_2 \pm c_1 e^{-\frac{x}{\lambda_{N B}}} + \pm c_2 e^{\frac{x}{\lambda_{N B}}} \tag{9}$$

for N and N_B respectively, where $b_1$, $b_2$, $c_1$, and $c_2$ complete the number of coefficients that describe the spin-dependent transport in the present trilayer system. The spin accumulation in each layer of the F[N]N_B nanowire are $\mu_N^x(x)$, while the charge chemical potentials read $\mu_C^x(x)$, and the spin-dependent current in a bulk ferromagnetic metal is (Ohm’s Law):

$$I_X^a(x) = -A_c \sigma_X^a \nabla \mu_X^a(x) / e \tag{10}$$

where $X = F, N, N_B$ and $\sigma_X^a = \sigma_N / 2$. The spin current $I_X^a = I_X^\uparrow - I_X^\downarrow$ is the difference between spin-up and spin-down currents.

where parameter such as $R_{\lambda_X} = \rho_X \lambda_X / A_c$, which is the resistance over the spin-flip diffusion length $\lambda_X$ in $X$ and $\rho_X$ is the corresponding electrical resistivity, are implicit in the calculations.

### A. Interface resistances

Next we consider spin-dependent transport through the interfaces. We disregard interface-induced spin-flips so at the F|N interface:

$$I_1^a = \frac{G_1^a}{e} [\mu_F^a(0) - \mu_N^a(0)] \tag{11}$$

where $G_1^a$ is the interface conductance with polarization $P_1 = (G_1^{\uparrow} - G_1^{\downarrow}) / G_1$ and $G_1 = G_1^{\uparrow} + G_1^{\downarrow}$. At the interface between the two normal metals N|N_B

$$I_2^a = \frac{G_2^a}{e} [\mu_N^a(L) - \mu_{N B}^a(L)] \tag{12}$$

Charge ($I_c = I_{1,2} = I_{1,2}^{\uparrow} + I_{1,2}^{\downarrow}$) and spin ($I_s^a = I_s^{\uparrow} - I_s^{\downarrow}$) currents are conserved in the interfaces 1 and 2, and assuming that $R_1 = 1/G_1$ and $R_2 = 1/G_2$.

### B. Boundary conditions

The boundary conditions are spin and charge current conservation at the interfaces.

$$I_F^a(0) = I_N^a(0) = I_2^a \tag{13}$$

for the F|N interface and

$$I_N^a(L) = I_{N B}^a (L) = I_2^a \tag{14}$$

for the N|N_B interface. We assume that the spin accumulation vanishes at the end of N_B

$$\mu_{N B}^a (L + A) = 0 \tag{15}$$

which is valid for $L_N \gg \lambda_N$ or $L_B \gg \lambda_{N B}$ and/or when nanopillar diameter widens at $L_B$. We can now determine $\beta_1$, $b_1$, $b_2$, $c_1$, and $c_2$ in terms of the coefficients.

Then, it can be now computed the spin accumulation, spin current and charge electrical potential.

The total electrical resistance, $R = \mu_c / (e I_c)$, of the device can now be written as

$$R = R_{F|N|N_B} = R_F(x = -L_F) - R_{N_B}(x = L + L_B) \tag{16}$$

$$R(T_0) = -\frac{2P_F}{I_c(1 - P_F)} \beta_1 e^{L_F / \lambda_X} + \frac{\alpha_1}{I_c} \rho_F L_F / A_c \tag{17}$$

where

$$\alpha_1 = I_c R_1 - \frac{2\beta_1 (P_1 - P_F)}{1 - P_F^2} + P_1 (b_1 + b_2) \tag{18}$$

and

$$\alpha_2 = -I_c R_2 - \frac{I_c \rho_{N L}}{A_c} \tag{19}$$

are the voltage drop at the two interfaces.

Numerical results for the transport properties require the parameters of the samples considered by Bosu et al. at room temperature $T_B$. The ferromagnetic metal is typically a Heusler alloy Co$_2$MnSi (CMS) while the normal metal N is gold and N_B is Cu. The resistivities and spin-flip diffusion lengths are given in Table I.

### Table I. Spin-flip diffusion length and electrical resistivity at 300K used for the F|N|N_B nanopillar structure

| Material   | $\lambda$ (nm) | $\rho$ (\(\mu\Omega\) cm) |
|------------|----------------|---------------------------|
| Co$_2$MnSi | 2.1            | 70.0                      |
| Au         | 60             | 2.27                      |
| Cu         | 350            | 1.73                      |

Fig. 2 illustrates that a charge current $I_c$ leads to a spin accumulation over the spin-flip diffusion length $\lambda_F$. 

\[ \text{Fig. 2} \]
in F, reaching its maximum value at the F\|N interface, where the spin is injected\textsuperscript{26,27,29,30} and decays exponentially along the spin-flip diffusion length of the normal metals $\lambda_{N,N_B}$. The spin current is plotted in Fig. 3. In a normal metal the spin current is proportional to the gradient of the spin accumulation, Eq. (10). It is observed in the model (Fig. 3) that the spin current decays rapidly in the central island N. Its behavior depends strongly on the spin-flip diffusion length of the metal, for our model we have $\lambda_{Au} < \lambda_{Cu}$. Additionally, it has an influence from the design length $L, L_B$ and the boundary condition established in Eq. (15).

### III. SPIN-DEPENDENT THERMOELECTRICITY OF F\|N\|N_B PILLOWS

In the experiments the electrical resistance change is measured as a function of applied current, reflecting the balance between the Joule heating and Peltier cooling. In order to model this effect we need to compute the temperature profile distribution $T(x)$ over F\|N\|N_B pillars. Temperature distributions have been previously calculated, but without taking Joule heating into account in spin-dependent systems\textsuperscript{26} Assuming that we know the temperature dependence of the electrical resistivity $\rho(T)$ and interface resistances $R_{1,2}(T)$, the total temperature dependent resistance reads

$$\Delta R = \frac{1}{T} \int R(T(x)) \, dx - R(T_0) \quad (20)$$

where $R(T_0)$ is given in Eq. (17). For simplicity, we disregard the heat leaked through the cladding of the nanopillar, which is valid when the thermal contact is weak or the cladding material has a much smaller heat conductivity. Significant heat leakage would reduce the temperature gradients calculated here, leading to an overestimate of the thermoelectric cooling power. In the following we determine the heat current and its divergence in the nanopillar taking into account the Kapitza thermal resistances at interfaces\textsuperscript{26} The temperature profile distribution along the nanopillar structure is calculated using heat conservation at interfaces, to finally describe the performance of the nanodevice in the resistance-current (R-I) characteristics. Except for the temperature dependence of the resistance that serves as a thermometer, we disregard the (for elemental metals) weak temperature dependence of the thermoelectric parameters.

In the Sommerfeld approximation the linear response relations between currents and forces in bulk materials read\textsuperscript{26}

$$\begin{pmatrix} J_c \\ J_s \\ J_q \end{pmatrix} = \sigma \begin{pmatrix} 1 & P_F & ST \\ P_F^{-1} & 1 & P_F^{ST} \\ ST/P_F & P_F^{ST} & \kappa \tau / \sigma \end{pmatrix} \begin{pmatrix} -\partial_x \mu_c / e \\ -\partial_x \mu_s / (2e) \\ -\partial_x \ln \tau \end{pmatrix}$$

(21)

where $S$ is the (charge) Seebeck coefficient, $\sigma$ the electrical conductivity, $\kappa$ the thermal conductivity, all at the Fermi energy and $T$ is the temperature (disregarding spin temperatures\textsuperscript{35}). Here, $J_c = I_c / A_c$, etc., are current densities.

$$P'_F = \frac{\partial}{\partial E} \left( \sigma^F_P - \sigma^F_F \right) E_F$$

(22)

is the spin polarization of the energy derivative of the conductivity at the Fermi energy, which is related to the spin polarization of the thermopower as

$$P_S = \frac{S_T - S_L}{S_T + S_L} = \frac{P'_F - P_F}{1 + P'_F P_F} \quad (23)$$
Joule heating is a source term that causes a divergence in the heat current:

\[
\frac{\partial}{\partial x} J_q = -J_c \frac{\partial}{\partial x} \frac{\mu_c}{e} \tag{24}
\]

Including the dissipation due to spin relaxation, we obtain the matrix expression for the divergence of the current densities

\[
\frac{\partial}{\partial x} \begin{pmatrix}
J_c \\
J_q
\end{pmatrix} = \begin{pmatrix}
0 & 0 \\
-P_F & -2P_F^2 \frac{2\rho F}{\mu_c} & 0
\end{pmatrix} \begin{pmatrix}
\mu_c \\
\frac{\mu_c}{e} T
\end{pmatrix}.
\tag{25}
\]

### A. Heat currents and temperature profiles in the bulk of the layers

The divergence of the heat current in the ferromagnet F reads (Eq. (25))

\[
\frac{\partial}{\partial x} J_q^F = J_c^2 \rho_F + \frac{(1 - P_F^2) \mu_s^2}{4 \rho_F \lambda^2} + J_q^2 \frac{\mu_s}{\kappa} \tag{26}
\]

which equals the derivative of the heat current in Eq. (24)

\[
\frac{\partial}{\partial x} J_q = \frac{\partial}{\partial x} \left( J_c S_F T - \frac{(P_F - P_F^2) S_F T \mu_s}{2 \rho_F \lambda} - \kappa_F \frac{\partial}{\partial x} T \right) \tag{27}
\]

\[
= -\frac{(P_F - P_F^2) S_F T \mu_s}{\rho_F \lambda^2} - \kappa_F \frac{\partial^2}{\partial x^2} T, \tag{28}
\]

leading to the heat diffusion equation

\[
\frac{\partial^2}{\partial x^2} T = -\frac{(P_F - P_F^2) S_F T \mu_s}{\rho \kappa} - \frac{J_c^2 \rho_F^2}{2 \lambda^2} - \frac{(1 - P_F^2) \mu_s^2}{4 \rho \kappa \lambda^2} - \frac{J_q^2}{\kappa^2} \tag{29}
\]

Heat transport is carried in parallel by phonons and electrons. We assume here efficient thermalization in and between both subsystems, meaning that the electron and phonon temperatures are taken to be identical. The total thermal conductivity then reads \(\kappa = \kappa_e + \kappa_p\).

For the ferromagnetic metal F we set \(T(x = 0) = T_{N1}\) where \(T_{N1}\) is governed by the boundary condition at the F/N interface discussed in the next subsection, while \(T(x = -L_F) = T_L\) is fixed by the reservoir (see Fig. [3]). The solution of the heat diffusion Eq. (29) disregarding the generlized Thomson effect term \(-J_q^2/\kappa^2\) then becomes

\[
T_F(x) = -\frac{2(P_F' - P_F) S_F T_{N1} \beta_1}{\kappa_F \rho_F (1 - P_F^2)} \left[ e^{\frac{2\rho_F x^2}{\kappa_F A_e}} - 1 \right] - \frac{I_c^2 \rho_F x^2}{2 \kappa_F A_e} + \frac{\beta_1^2}{\kappa_F \rho_F (1 - P_F^2)} \left[ e^{\frac{2\rho_F x^2}{\kappa_F A_e}} - 1 \right] + T_{N1} \tag{30}
\]

and

\[
\frac{\partial}{\partial x} T_F = -\frac{2(P_F' - P_F) S_F T_{N1} \beta_1}{\kappa_F \rho_F (1 - P_F^2)} \left( e^{\frac{2\rho_F x^2}{\kappa_F A_e}} - 1 \right) - \frac{\beta_1^2}{\kappa_F \rho_F (1 - P_F^2)} \left( e^{\frac{2\rho_F x^2}{\kappa_F A_e}} - 1 \right)
\]

\[
+ \frac{I_c^2 \rho_F x^2}{\kappa_F A_e} \left( x - \frac{L_F}{2} \right) + \left( \frac{T_L - T_{N1}}{L_F} \right). \tag{31}
\]

Finally, the heat current distribution reads:

\[
J_q \cdot A_c = S_F T_I c e + \frac{2(P_F' - P_F) S_F T_{N1} \beta_1}{(1 - P_F^2)} \left( 1 - e^{\frac{2\rho_F x}{R_F}} \right)
\]

\[
+ \frac{\beta_1^2}{(1 - P_F^2)} \left( e^{\frac{2\rho_F x}{R_{\lambda_F}}} + 1 - e^{\frac{2\rho_F x}{R_F}} \right)
\]

\[
+ I_c^2 \left( \frac{R_F x}{L_F} - \frac{R_F}{2} \right) - \frac{\kappa_F A_e}{L_F} (T_L - T_{N1}). \tag{32}
\]

where \(R_F = \rho_F L_F/A_e\) is the electrical and \(R_{\lambda_F} = \rho_F \lambda_F / A_e\) the spin resistance.

Repeating this analysis for normal metals, we obtain a
heat current in N

\[ J_q^N(x) \cdot A_c = S_N T I_c + I_c^2 \left( R_N \frac{x - R_N}{L} - \frac{R_N}{2} \right) \]
\[- b_1^2 \left( \frac{e^{-2\frac{L}{R_N}}}{2R_{N^2}} + \frac{e^{-\frac{2L}{R_N}}}{4R_N} \right) \]
\[ + b_2^2 \left( \frac{e^{\frac{2L}{R_N}}}{2R_{N^2}} - \frac{e^{-\frac{2L}{R_N}}}{4R_N} \right) \]
\[ - \frac{b_1b_2}{R_{\lambda_N}} \left( 2x - \frac{L}{\lambda_N} \right) - \frac{\kappa_N A_c}{L} \left( T_{N2} - T'_{N1} \right) \]
\[ \text{(33)} \]

and Nb

\[ J_q^{NB}(x) \cdot A_c = S_{NB} T I_c + I_c^2 \rho_{NB} \left( x - \frac{2L + L_B}{2} \right) \]
\[- c_1^2 \left( e^{-2\frac{L}{R_{NB}}} 2R_{NB} + \frac{e^{-\frac{2L}{R_{NB}}}}{4R_{NB}} \right) \]
\[ + c_2^2 \left( e^{2\frac{L}{R_{NB}}} 2R_{NB} - \frac{e^{-\frac{2L}{R_{NB}}}}{4R_{NB}} \right) \]
\[ - \frac{c_1c_2}{R_{\lambda_{NB}}} \left( 2x - \frac{2L + L_B}{\lambda_{NB}} \right) - \frac{\kappa_{NB} A_c}{L_B} \left( T_R - T'_{N2} \right) \]
\[ \text{(34)} \]

B. Interfaces

Finally, we knit the solutions for the bulk layers together at the interfaces by boundary conditions. The contacts to an abruptly widening nanopillar may be treated as ideal reservoirs (heat and spin sinks) at constant temperatures \( T_L = T_R = T_0 \) (see Fig. 1). By disregarding interface-induced spin-flips and, for the moment, the Joule heating by the interface resistance, we may impose charge, spin and energy conservation at each interface \[11,40,41\] such as

\[ J_q^N(x = 0) = J_q^{NB}(x = 0) \]
for F[N, where analogous to Eq. \[21\] \[24\].

\[ J_q^1 \cdot A_c = G_{TH1} A_c \Delta T - G_1 S_1 T_1 \Delta \mu_{c(1)}^{(1)} - P' F G_1 S_1 T_1 \frac{\Delta \mu_{s(1)}^{(1)}}{2} \]
\[ \text{(35)} \]
is the interface heat current, \( G_{TH1} \) the Kapitza thermal conductance (including the phonon contribution), \( A_c \) the cross sectional area of the nanopillar, \( \Delta T = T_{N1} - T_{N1}' \) the temperature drop over the interface, \( T_1 = (T_{N1} + T_{N1}')/2 \) the interface temperature, \( G_1 \) the electrical interface conductance, \( S_1 \) the interface thermopower, and \( \Delta \mu_{c(1)}^{(1)} \) the charge (spin) accumulation differences over the interface.

Substituting Eqs. \[32\] and \[33\] for \( x = 0 \) leads to

\[ T_{N1} = \left\{ \begin{array}{ll}
- (S_N I_c + \frac{\kappa_N A_c}{L}) & - \frac{\kappa_F A_c T_L}{H_2 L_F} - \frac{I_c^2 R_F}{2H_2} \\
+ \frac{\beta_1^2}{H_2(1 - P'_F)} & \left( \frac{2}{R_{\lambda_F}} - \frac{e^{\frac{2L'}{R_F}}}{R_F} - 1 \right) \\
- \frac{I_c^2}{2} (R_F - R_N) & - \frac{\kappa_F A_c T_L}{L_F} + \frac{\kappa_N A_c T_{N2}}{L} \\
+ \frac{\beta_1^2}{(1 - P'_F)^2} & \left( \frac{2}{R_{\lambda_F}} - \frac{e^{\frac{2L'}{R_F}}}{R_F} - 1 \right) \\
b_1^2 \left( \frac{1}{2R_{\lambda_N}} + \frac{\kappa_{NB} A_c}{L} \right) & - \frac{b_1b_2}{R_{\lambda_N}} \left( \frac{e^{-\frac{L}{\lambda_N}}}{4R_N} \right) \\
- \frac{2}{(1 - P'_F)} & \left( \frac{2}{R_{\lambda_N}} - \frac{e^{\frac{L}{\lambda_N}}}{R_F} - 1 \right) \\
+ \left( \kappa_{NB} A_c \right) & - \frac{b_2}{R_{\lambda_N}} \left( \frac{e^{-\frac{L}{\lambda_N}}}{4R_N} \right) \\
- \frac{2}{(1 - P'_F)} & \left( \frac{e^{-\frac{L}{\lambda_N}}}{R_F} - 1 \right) \\
\left( \frac{S_N I_c + \frac{\kappa_N A_c}{L}}{H_2} + \frac{\kappa_F A_c}{H_2 L_F} + \frac{2(P'_F - P_F)S_F \beta_1}{H_2(1 - P'_F)^2} \right) & \left( \frac{e^{\frac{L}{\lambda_N}} - 1}{R_F} \right) - \frac{H_1}{H_2} \\
- \frac{S_F I_c}{H_2} & \left( \frac{e^{\frac{L}{\lambda_N}} - 1}{R_F} \right) \}
\text{(36)} \]

and

\[ T'_{N1} = \left( \frac{S_F I_c}{H_2} + \frac{\kappa_F A_c}{H_2 L_F} + \frac{2(P'_F - P_F)S_F \beta_1}{H_2(1 - P'_F)^2} \right) \]
\[ \left( \frac{e^{\frac{L}{\lambda_N}} - 1}{R_F} \right) - \frac{H_1}{H_2} \]
\[ - \frac{\kappa_F A_c T_L}{H_2 L_F} - \frac{I_c^2 R_F}{2H_2} + \frac{\beta_1^2}{H_2(1 - P'_F)^2} \]
\[ \left( \frac{2}{R_{\lambda_F}} - \frac{e^{\frac{L}{\lambda_N}}}{R_F} - 1 \right) \]
\[ \text{(37)} \]

where \( H_1(2) = -G_1 S_1 \Delta \mu_{c(1)}^{(1)}/2 - P'_F G_1 S_1 \Delta \mu_{s(1)}^{(1)}/4 \pm G_{TH1} A_c \). We may determine the temperatures \( T_{N2} \) and \( T_{N2}' \) at interface N[N_B] analogously.

Eqs. \[36\] and \[37\] include bulk and interfacial Peltier effects as well as Joule heating in the bulk materials (see Fig. 1) but not yet the interfacial Joule heating. Here we focus on Joule heating by the N[N_B] interface, which is the dirty one in existing experiments. We can treat interface heating easily in two limiting cases. In the dirty limit the interface is a resistor with small but finite thickness \( L_I \) around the position \( x = d_I \) in which the electrons
dissipate their energy directly to the lattice:

$$\frac{\partial}{\partial x} J_q^{c} = \left\{ \begin{array}{ll} \frac{J_{c} R_{1} A}{0} & \text{for } -L_{1}/2 < x - d_{1} < L_{1}/2 \\ 0 & \text{otherwise} \end{array} \right.$$  (38)

Clean interfaces, point contacts or coherent tunnel junctions, on the other hand, inject hot electrons (and holes) into the neighboring layers where they lose their excess energy on the scale of the electron-phonon thermalization length $\lambda_{ep}$. In normal metals like Cu it is surprisingly large even at room temperature, i.e. $\lambda_{ep}^{c} \approx 60 \text{ nm}$.

The clean limit (assuming that $\lambda_{ep}^{c} < \lambda_{ep}^{b}$ is smaller than the pillar length)

$$\frac{\partial}{\partial x} J_q^{c} = \left\{ \begin{array}{ll} \frac{J_{c}^{2} R_{1} A}{0} \lambda^{c}_{ep} & \text{for } -\lambda^{c}_{ep} < x - d_{1} < \lambda^{c}_{ep} \\ 0 & \text{otherwise} \end{array} \right.$$  (39)

The two limits therefore differ only by the volume in which the heat is produced. In the extreme case of $\lambda_{ep}^{c} \gg L_{N}$ all interface Joule heating occurs in the reservoirs, where its effect can be disregarded. In the following we consider both extremes, i.e. the dissipation occurs either in the interfacial thickness $L_{1}$ or in the reservoirs $\lambda_{ep}^{c} + \lambda_{ep}^{b} = \infty$.

We can implement these models into Eqs. (30) and (37) as follows. In Eqs. (38) and (39), Joule heating is represented by the power density $\frac{J_{c}^{2} R_{1} A}{0}$ in the volume $V = AL_{1}$. The total power dissipated at the interface is therefore $\frac{J_{c}^{2} R_{1} A}{0}$. This term can be added to Eq. (36): the first term of the third line expresses the balance between the Joule heating of the bulk metals to which the interface contribution may be added. The interfacial Joule heating thereby reduces the cooling power of the nanopillar. By contrast, in the ballistic limit and long relaxation lengths Joule heating is deferred to the heat sinks, and does not contribute at all. In Eq. (37) the interfacial Joule heating is indirectly related by the already determined term $T_{N1}$ of Eq. (36). A regular sequence of the Joule heating is represented by a parabola-like curve, but the interfacial resistance is a factor of temperature behaviour to result in a small kink in the temperature distribution at the interface which is interpreted as bulk heating to be dominant in comparison with the interfacial one.

IV. RESULTS

In general, interfacial resistances $R_{1/2}$ may vary from close to zero for good metallic contacts to that of a very thin (Ohmic) tunnel barrier. A highly resistive interface can, e.g., be caused by a sample fabrication process in which the vacuum is broken, leading to organic deposits. We simulate resistive F|N or N|N$_B$ interfaces by modulating $R_{1/2}$ from zero resistance to a large value. A large resistance of either interface turns out to enhance the cooling effect as long as the interfacial Joule heating does not dominate, i.e., when the current bias is not too large.

A. Temperature profiles in a F|N|N$_B$ pillar

We are interested in the temperature profile in a pillar with equal temperatures of the two external reservoirs $T_L = T_R = T_0$, noting that the model can be easily extended to calculate the thermopower due to a global temperature difference over the device. We start with $T_{N1} = T_{N2} = T_{N3} = T_0$ as initial conditions (see Fig. 1), which is substituted into Eqs. (36) and (37) to obtain the first iteration. The temperature profiles converge after several iterations.

Results for F|N|N$_B$ nanopillars are shown in Fig. 5 for different current densities, with temperature $T_0$ in the reservoirs maintained at 300K, using parameters from Tables I - III for bulk and interfaces, for the case of all Joule heating occurring in the reservoirs. The top panel of Fig. 5 is for clean interfaces with $A_{c}R_{1} = 0.915 \text{ fΩm}^{2}$ and $A_{c}R_{2} = 0.34 \text{ fΩm}^{2}$. Values of interfacial electrical resistance are well-known parameters, while those of Kapitza heat conductance are not, specially for F|N interfaces, and value of Kapitza heat conductance in latter is assumed not to be such a good heat conductor as compared with the second interface. The Joule heating is generated mainly by the relatively resistive ferromagnet, while the cooling takes place at the F|N interface, giving rise to a complex temperature and heat current distribution. The dotted lines for each curve show the average temperature in the different layers $T_{INT}$ that govern the resistance change of the pillar.

The bottom panel of Fig. 5 shows the temperature profile in the presence of a dirty interface N|N$_B$ with a 100 times larger resistance $A_{c}R_{2} = 34 \text{ fΩm}^{2}$. $G_{TH,2} = 5.9 \cdot 10^{7} \text{ W/m}^{2}\text{K}$ is assumed to be reduced by the same ratio, while other parameters are kept the same. The dissipation at the dirty interface N|N$_B$ locally increases the temperature in the normal metals. A marked discontinuity of the temperature at N|N$_B$ interface develops due to the small thermal conductance $G_{TH,2}$. The temperature on the F-side drops from approximately 298.3 K for a clean N|N$_B$ interface to 297.7 K for the dirty one (see Fig. 5). The increased interface resistance forms a barrier for the heat flow from the heat sinks towards the interface, allowing the region close to the interface to cool down more efficiently, thereby enhancing the effective Peltier effect.

B. Peltier cooling, Joule heating, and R-I characteristics

According to Eq. (20) the temperature profile $T(x)$ is directly related to the observable resistance change. We compute a specific temperature profile for a given current bias as sketched below, which can be used to obtain the total resistance as a function of current that may be compared with experimental results. To this end
TABLE II. Thermoelectric parameters of the bulk metal layers in the F[N][N]$_B$$_2$ nanopillars at 300 K: Thermal conductivities $\kappa$ (W/mK), and Seebeck coefficients $S$ ($\mu$V/K). $P_F$ is the polarization of the conductivity for the ferromagnet while $P_F'$ is the polarization of its energy derivative. For lack of sufficient data we take $P'_F = P_F = 0$, thereby disregarding much of the spin-dependence of the heat diffusion equations. $R_X (L_X)$ are the resistances in $\Omega$ when thicknesses of the metal layers are in $\mu$m.

| Material | $\kappa$ | $S$ | $P_F$ | $\partial R_X / \partial T$ |
|----------|---------|-----|-------|---------------------------|
| Co$_2$MnSi | 15 | -20 | 0.71 | $6.07 \times 10^{-15} \cdot (L_F / A_c)$ |
| Au (N) | 318 | 1.83 | 8.14 $\times 10^{-11} \cdot (L/A_c)$ |
| Cu (N$_B$) | 401 | 1.94 | $6.84 \times 10^{-11} \cdot (L_B/A_c)$ |

TABLE III. Interfacial thermoelectric parameters of the F[N][N]$_B$$_2$ nanopillars at 300 K. Interface Kapitza thermal conductances $G_{TH}$ (W/m$^2$K) including the phonon contribution. $S$ ($\mu$V/K) is the interfacial Seebeck coefficient and $P$ the spin polarization of the interface conductance.

| Material | $G_{TH}$ | $S$ | $P$ | $\partial R_X / \partial T$ |
|----------|---------|-----|-----|---------------------------|
| CMS/Au | $1.8 \times 10^9$ | -4 | 0.77 | $(\partial R_F / \partial T + \partial R_F' / \partial T)/2$ |
| Au/Cu | $5.9 \times 10^9$ | 3.5 | 0 | $(\partial R_N / \partial T + \partial R_{N_B} / \partial T)/2$ |

we linearize Eq. (20) as:

$$\Delta R_X \approx \frac{\partial R_X}{\partial T} (T_{X_{AVG}} - T_0).$$

(40)

The total resistance differential is governed by the temperature dependence of the layer and interface resistances. Each bulk material layer has a specific $\partial R_X / \partial T$, while the calculations establish average temperatures $T_{X_{AVG}}$ for the sections F, N, and N$_B$ respectively, as shown in Fig. 5 marked by dotted lines. Highly resistive interfaces may affect or even dominate the global resistance change when $R_1(2)$ and $\partial R_1(2) / \partial T$ are large. Our calculations include the temperatures at interfaces $T_{1(2)}$ as expressed in Eq. (53). For the temperature dependence of the bulk resistivities we adopt the values listed in Table II. For resistive interfaces we average $\partial R_X / \partial T$ of the two materials; This is expressed in N[N]$_B$ interface as represented in Table III.

$$\frac{\partial R_2}{\partial T} = \frac{1}{2} \left( \frac{\partial R_N}{\partial T} + \frac{\partial R_{N_B}}{\partial T} \right),$$

(41)

while we disregard the temperature dependence of the resistance for good interfaces.

In Fig. 6 the effect of inserting a highly resistive N[N]$_B$ interface on the R-I curves is shown for the scenario when the interface Joule heating is very non-local, i.e. use Eq. (53). The (effective) Peltier cooling (blue line, bottom) is visibly enhanced. The change in the total resistance can be understood in terms of the temperature distribution along the pillar as shown in Fig. 5. The increased interfacial resistance $R_2$ improves the effective Peltier coefficient $\Pi$ from $11.2$ mV for a clean interface to $\Pi = 23.9$ mV in the case of a dirty interface. Additionally, a change in the Peltier coefficient from $\Pi = 23.9$ mV to $\Pi = 24$ mV is reached when Eq. (41) is implemented into this computation. We should note that while the effective Peltier coefficient is enhanced by a highly resistive interface under a constant current bias, it becomes a more efficient system, viz. the nanopillar requires a lower applied voltage in combination with more cooling effect simultaneously.

C. Trilayer nanopillar model

We now evaluate the thermoelectric performance as a function of structural and material parameters of the nanopillars. Matching Bosu et al.'s samples, we adopt bulk (Drude) thermopowers of the leads as $S_F = $
$S_{CMS} = -20 \mu V/K$ for the ferromagnetic Heusler alloy (Co$_2$MnSi$_{0.57}$Fe$_{0.43}$), $S_{Au} = 1.83 \mu V/K$ for the normal metal N and $S_{Cu} = 1.94 \mu V/K$ in normal metal $N_B$.

Our model is scale-invariant with respect to the pillar diameter, so we cannot explain the enhanced effective Peltier cooling found in the narrowest pillars by the experiment in terms of an intrinsic size effect. However, smaller structures can be more susceptible to the effects of e.g. incomplete removal of resist material used during nanofabrication. We have discussed above that such extrinsic effects do affect the thermoelectric properties and can be treated in our model. The interfacial thermopower $S_{CMS|Au}$ and its spin polarization $P_S$ are basically unknown parameters that may contribute importantly to the cooling effect in nanostructures, as reflected in the enhancement of the global effective Peltier coefficient $\Pi = 11.2 mV$ for $S_1 = -4 \mu V/K$ to $23.2 mV$ for $S_1 = S_{CMS|Au} = -30 \mu V/K$; this case is especially relevant in the presence of a resistive $N|N_B$ interface.

The effects of an enhanced interface resistance $A_e R_1(2)$ on the Peltier cooling can also be tested by varying it from that of a good intermetallic to a value corresponding to a thin tunnel barrier. The interface resistance turns out to improvement of $\Pi$ as long as the additional Joule heating does not dominate, as illustrated in Fig. 6. Furthermore, in Fig. 7 it is plotted the temperature profile distribution when Joule heating is generated in the interfaces, setting the nanopillar with the same parameters of Fig. 5 except for the modulus of the electrical tunnel junction nor the Kapitza thermal conductance, in which both have the same ratio of change. It can be compared clearly a decrement in the performance of the cooling device for this case, since the Joule heating produced at the interface counteracts the cooling of Peltier effect. As discussed above, the interface resistance hinders the flow of heat current from the heat baths towards the cooling interface. For an interfacial resistance of $A_e R_1 = 0.915 f\Omega m^2$ and $A_e R_2 = 0.34 f\Omega m^2$, the total Peltier coefficient reaches a value of $\Pi_{CMS|Au|Cu} = 11.2 mV$, matching parameters from Tables II and III where this result from this theoretical model is close to experimental ones. A linear dependence of the Peltier coefficient was found when varying the interface resistance area $A_e R_1$ from 0.915, 9.15 and 91.5 (f$\Omega m^2$), resulting in Peltier coefficients $\Pi_{CMS|Au|Cu}$ of 11.2, 13.49, and 31.61 mV, respectively. By contrast, when the interfaces are clean and Joule heating is suppressed (assuming $\lambda_{ep}^{F} + \lambda_{ep}^{B} = \infty$), the Peltier coefficients increase to 11.28, 14 and 42 mV for the same interface resistances for the best case when Joule heating is all produced in the reservoirs.

Since our calculations take the spin degree of freedom into account the spin accumulations and spin currents along the nanopillar are byproducts of the calculations. In contrast to $|P_F| < 1$, the spin polarization of the derivative of the conductivity $\sim \lambda_{ep}^{F} < \infty$. When $P_F < P_F'$ the spin contribution to the cooling power is proportional to the spin accumulations as expressed in Eqs. (36) and Eq. (37). A Peltier coefficient of $11.2 mV$ with parameters from Tables II and III is increased by a factor 2 when $P_F' = -20$. However, when $P_F' > P_F$, the spin degree actually generates heating thereby reducing the cooling power.

We also studied the dependence of the effective cooling on layer thicknesses $L_F, L$ and $L_B$. The Joule heating dominates for a critical current bias $I_c$ that decreases with increasing $L_F$. When the thickness of F=CMS is
reduced from 40 nm to approximately 5 nm. II improves slightly from 11.2 to 12.5 mV. The optimal thickness of the ferromagnetic film is $L_F \sim \lambda_F$. The normal metals do not significantly contribute to the cooling since their Peltier coefficients are relatively small.

Finally, slight enhancements of the Peltier coefficient could be achieved by including in the analysis an external heat current $J_{ext}$, which is depicted in the left hand of Fig. 11, which forms part of an extension of the nanopillar that could lead the head current towards a further reservoir so that $T_H > T_0$, to result in a slight enhancement of the Peltier effect. This makes a more sophisticated model, but we leave it for a future study.

V. SUMMARY AND CONCLUSIONS

This paper is motivated by the observed enhancement of the cooling power in magnetic pillars when the cross section was reduced to the nanoscale. We develop a realistic spin, charge, and heat diffusion model to investigate the roles of spin-dependent bulk and interface scattering contributions. We analyzed the (apparent) cooling power and the conditions to maximize the effective Peltier effect.

We demonstrate that very thin (Ohmic) tunnel junctions can improve the cooling power of devices as apparent in the shift of $R(I)$ parabolas. On the other hand, the spin degree of freedom that was thought to be essential in CMS materials appears to be less important for conservatively chosen parameters. However, the material dependence of key parameters is basically unknown. The parameter $P_F$, i.e. the spin polarization of the spectral asymmetry of the conductance, turns out to play an important role. This parameter may become arbitrarily large when $\partial (\sigma^F_+ + \sigma^F_-) / \partial E \big|_{E_F} = 0$ or, for interfaces $\partial \left( G^F_+ + G^F_- \right) / \partial E \big|_{E_F} = 0$, which does not seem to be an exotic condition and we recommend a systematic search for such materials or material combinations. Our results also indicate that interfacial parameters such as the interface Seebeck coefficients $S_{1(2)}$ play a very significant role in the thermoelectric characteristics of multilayers and may not be disregarded when validating their performance and in agreement with Hu et al. affirming that the value of this coefficient is even larger that the conventional one, that in the present model both contribute simultaneously in the cooling effect.

While the experiments up now have been analyzed in a simplistic model for the compensation current at which heating and cooling effects cancel, we established a distributed model of currents and temperatures. The computed temperature profiles along the nanopillar established that the cooling is not homogeneous, but heating and cooling coexist in different locations of the sample. The current-dependent resistance only a very crude thermometer that is not a reliable measure for a cooling power that could be of practical use.

We find that it is possible to selectively cool a ferromagnet by a few degrees simply by a current flow in the right direction. This could be an important design parameter for STT-MRAMs. The writing of a bit of information by a switching event of the free layer in a memory element is accompanied by significant Joule and Gilbert heating. Applying a small bias current after the magnetization reversal can assist a quick return to the ambient temperature.

Our model is scale invariant with respect to the pillar diameter and does not provide and intrinsic mechanism for the observed size dependence of the Peltier effect. In principle, extrinsic effects should exist. The large fluctuations observed in the experimental results indicates significant disorder in the smallest nanopillars. One source of the problems can be the need to break the vacuum during sample fabrication. The effect of pollutants at an interface are then likely to be more serious for smaller pillars. We found indeed that by modelling interface as a thin tunnel junction enhances the apparent Peltier coefficients by suppressing the heat currents flowing into the pillar from the reservoirs. However, the record cooling effects observed for some of the narrowest pillars appear to be beyond the effects that can credibly be modelled, and we cannot exclude the possibility that something more interesting is going on.

Several effects are beyond the present model approach. Size quantization is not expected to be important in metallic structures at room temperature, but could play a role in heterogeneous materials disordered on a nanometer-scale. Spin waves and magnons, i.e. excitations of the magnetic order parameter, affect thermoelectric properties. The magnon-drag effect enhances the Seebeck coefficient. The longitudinal spin Peltier effect discovered for bilayers with magnetic insulators should also exist in metallic structures: the spin accumulation in the normal metal generates a heat current that comes on top of the heat currents discussed here. It is not clear, however, how and why these effects become so strongly enhanced in the nanopillars addressed experimentally. More experiments on even smaller and more reproducibly fabricated nanopillars, preferably fabricated without breaking the vacuum, are necessary in order to provide hints on what is going on.

We conclude that the Peltier effect in magnetic nanopillars with diameters $\geq 100$ nm appears to be well understood, but that the enhanced values for narrower ones are to date only partly explained. In order to employ the large observed effects, more experiments are necessary in order to shed light on the underlying physical mechanisms.

ACKNOWLEDGMENTS

I. J. A. is grateful to O. Tretiakov, T. Chiba and A. Cahaya for fruitful discussions and all members of the Bauer Laboratory at the IMR, Tohoku University for their hos-
