Direct observation of the spin-dependent Peltier effect

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The Peltier coefficient describes the amount of heat that is carried by an electrical current when it passes through a material. When two materials with different Peltier coefficients are placed in contact with one another, the Peltier effect causes a net flow of heat either towards or away from the interface between them. Spintronics describes the transport of electric charge and spin angular momentum by separate spin-up and spin-down channels in a device. The observation that spin-up and spin-down charge transport channels are able to transport heat independently has raised the possibility that spin currents could be used to heat or cool the interface between materials with different spin-dependent Peltier coefficients. Here, we report the direct observation of the heating and cooling of an interface by a spin current. We demonstrate this spin-dependent Peltier effect in a spin-valve pillar structure that consists of two ferromagnetic layers separated by a non-ferromagnetic metal. Using a three-dimensional finite-element model, we extract spin-dependent Peltier coefficients in the range −0.9 to −1.3 mV for permalloy. The magnetic control of heat flow could prove useful for the cooling of nanoscale electronic components or devices.

Spin caloritronics is a new field that combines concepts from thermoelectricity, such as the Peltier effect, and spintronics. The coupling of heat transport with spintronics has generated novel ideas such as innovative spin sources, thermal spin-transfer torque, magnetic heat valves, Seebeck effects in magnetic tunnel junctions, and magnetically switchable cooling. In particular, pioneering experiments by Gravier et al. on multiple cobalt/copper multilayer nanowires have indicated the existence of spin-dependent Peltier coefficients, based on magnetothermogalvanic voltage measurements. However, direct experimental evidence for cooling or heating by spin currents has not been reported.

The spin-dependent Peltier effect is based on the ability of the spin-up and spin-down channels to transport heat independently. Figure 1a gives a schematic presentation of this concept when a pure spin current \( J_{\uparrow} = -J_{\downarrow} \) passes through a non-magnetic metal/ferromagnetic metal (N/F) interface. The associated Peltier heat current \( Q_{\Pi} \) is the sum of that of the two spin channels. As these travel in opposite directions (the charge flow is zero), a net heat flow will only arise if the amount of heat carried by the separate spin species is different. The Peltier coefficients for majority and minority electrons, defined as \( \Pi_{\uparrow,\downarrow} = Q_{\Pi_{\uparrow,\downarrow}}/J_{\uparrow,\downarrow} \), represent the amount of heat carried by the individual spin channels. In N, both coefficients are equal \( \Pi_{\uparrow} = \Pi_{\downarrow} \). However, in F, the spin-dependent Peltier coefficient, defined as \( \Pi_s = \Pi_{\uparrow} - \Pi_{\downarrow} \), is expected to be non-zero. Owing to spin flip processes, the spin current attenuates in the ferromagnet and within a few spin relaxation lengths \( \lambda_s \) from the interface, the Peltier heat current vanishes. Consequently, heat is effectively transferred from the interface into the ferromagnetic region over a finite length (or vice versa), thereby producing a temperature gradient (depicted in Fig. 1b) and a temperature drop \( \Delta T \).

To demonstrate the effect experimentally, a pure spin current is not required, but it can be accompanied by a charge current \( I_C = J_{\uparrow} + J_{\downarrow} \). To calculate the temperature gradient, the total heat current \( Q = Q_{\Pi} - \kappa VT \) is evaluated, where \( \kappa \) is the thermal conductivity of the electron and phonon systems and \( Q_{\Pi} = \Pi_{\uparrow} J_{\uparrow} + \Pi_{\downarrow} J_{\downarrow} \) is the Peltier heat current. If we assume that no heat can enter or leave the stack \( (Q = 0) \) and disregard Joule heating, the temperature gradient can be expressed as the sum of the charge and spin part of the Peltier effect (Supplementary Section A). For the spin part, the induced temperature difference between the F/N interface and the bulk of the ferromagnet is given by

\[
\Delta T = \frac{\alpha}{4\kappa} (1 - P_s^2) \Pi_s \mu_s^0
\]

where \( \alpha = \alpha_\uparrow + \alpha_\downarrow \) is the conductivity, \( P_s = (\sigma_\uparrow - \sigma_\downarrow)/(\sigma_\uparrow + \sigma_\downarrow) \) is the conductivity polarization, and \( \mu_s^0 = \mu_\uparrow^0 - \mu_\downarrow^0 \) is the spin accumulation at the interface. Therefore, we find that the induced temperature drop depends directly on the spin accumulation at the F/N interface.

The device used to study the spin-dependent Peltier effect consists of a stack of two ferromagnetic layers separated by a layer of N (Fig. 2). Assuming that the spin relaxation length \( \lambda_s \) is much larger than the thickness of N, we can neglect spin relaxation in N. A charge current \( I_C \) is sent through the stack and the temperature at the top is anchored at \( T_0 \). When the ferromagnets are aligned parallel (P), the spin current is constant over the whole stack and there

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Figure 2 | Spin-dependent Peltier effect in an F/N/F spin-valve stack. Schematics showing the spin electrochemical potential and temperature throughout the stack for parallel (P) and antiparallel (AP) configurations of the ferromagnets. Note that the splitting between \( \mu_+ \) and \( \mu_- \) and the heat profile at the F/N interfaces have been exaggerated for clarity (see Supplementary Section B for the actual electrochemical potential profile extracted from modelling). Spin-up/spin-down is defined as the spin direction of the majority/minority electrons in F2. a, In the P configuration, the spin current in the bulk of both ferromagnets equals the spin current in N, and no non-equilibrium spin accumulation exists. Hence, there is no spin-dependent Peltier effect. b, For the AP configuration, the bulk spin currents in F1 and F2 are opposite. The resulting spin accumulation at the F/N interface leads to a spin-dependent Peltier contribution and therefore an altered temperature gradient. c, The spin-dependent Peltier effect causes the temperature at the bottom contact to change between the P and AP alignments.

There is no non-equilibrium spin accumulation; that is, \( \mu_0 = 0 \) everywhere. The temperature follows a zigzag pattern, caused by the conventional (charge) Peltier effect (Fig. 2a). In the antiparallel (AP) alignment, the situation is different. Now the spin current in the bulk of F1 is opposite to the spin current in the bulk of F2, leading to a spin accumulation at the interfaces. According to equation (1), this gives rise to an additional temperature difference \( \Delta T \) (Fig. 2b) for each F/N interface in the stack. Hence, the induced difference in temperature at the bottom layer between both magnetic configurations is now \( \Delta T \) (Fig. 2c).

The specifically designed device was used to study the spin-dependent Peltier effect depicted in Fig. 2. It consists of a permalloy (Ni_{80}Fe_{20})(Py)/copper/Py spin valve stack (thickness, 15 nm/15 nm/15 nm; cross-section, 150 \( \times \) 80 nm²) with a platinum bottom contact and a gold top contact. Crosslinked polymethyl methacrylate (PMMA) was used for the insulating layer between these two contacts, forcing the applied current through the spin valve stack. To probe the temperature of the device a constantan (Ni_{45}Cu_{55})–platinum thermocouple was used, with constantan having been chosen because of its large Seebeck coefficient (~32 \( \mu V \) K\(^{-1} \); Supplementary Table 1 in Supplementary Section F). The thermocouple was electrically isolated from the bottom contact by an 8-nm-thick aluminium-oxide layer, thereby excluding any spurious voltage pick-up. During the measurement, a current was sent from contact 1 to contact 2 through the stack, while recording the thermovoltage between contacts 3 and 4. For the four-point spin-valve signal measurements, contacts 5 and 6 were used to probe the voltage. Using an a.c. lock-in measurement technique, it was possible to separate the Peltier contribution (\( \Delta T \propto I \)) and the Joule heating contribution (\( \Delta T \propto I^2 \)) by taking the first-harmonic \( V^{(1)} \) and second-harmonic \( V^{(2)} \) responses, respectively. Measurements were performed at room temperature.

The thermovoltage was recorded while sweeping the magnetic field in the in-plane direction from negative to positive and back. Figure 4a shows the first-harmonic response data with \( R_{3–4}^{(1)} \) defined as \( V^{(1)}_{3–4} / I \). The data show four abrupt changes in \( R_{3–4}^{(1)} \) when the magnetization of the Py layers switches from P to AP and back. A spin signal, \( R_{3–4}^{(2)} = (R_{3–4}^{(1)})^2 / I^2 \), of ~80 \( \mu \Omega \) is observed on top of a background signal, \( (R_{3–4}^{(1)})^2 / I \), of ~0.44 m\( \Omega \). This corresponds to a temperature difference at the thermocouple between the P and AP alignments of ~3 mK at 1 mA. Using the modelling described below, we calculated that \( \Delta T \approx 7.6 \) mK (equation (1)). The second-harmonic data are presented in Fig. 4b, where \( R_{3–4}^{(2)} = V^{(2)}_{3–4} / I^2 \), giving a spin signal of 110 mV A\(^{-2} \) on a background of ~1.73 V A\(^{-2} \). This signal originates from Joule heating in the device and its change between the P and AP configuration (Supplementary Section C).

The four-probe spin valve signal shown in Fig. 4c gives a ~100 mV spin signal. By matching the spin-valve signal from our three-dimensional finite-element model to this measured value, we obtain a conductivity polarization \( (P_\sigma) \) of 0.61, which is close to the bulk value for Py. The same model can now be used to extract the spin-dependent Peltier coefficient, \( \Pi_\Pi = \Pi_\Pi - \Pi_\Pi \), from the first-harmonic measurement in Fig. 4a. The previously obtained value for \( P_\sigma \), together with the electrical conductivities, thermal conductivities, spin relaxation lengths, Peltier coefficients and Seebeck coefficients for each material, are taken as input parameters (Supplementary Table 1 in Supplementary Section F).

From the spin-dependent Peltier signal in Fig. 4a, we then obtain a spin-dependent Peltier coefficient for Py of \( \Pi_\Pi = -0.9 \) mV. Similar results were obtained in two other devices, giving values for the spin-dependent Peltier coefficient of ~1.1 and ~1.3 mV (Supplementary Section D). The Thomson–Onsager relation applied to the separate spin channels \( \Pi_{\uparrow, \downarrow} = S_{\uparrow, \downarrow} T \) gives us a way

Figure 3 | Device geometry. a. Scanning electron microscopy image of the measured device. Yellow, gold contact; grey, platinum bottom contacts; blue, crosslinked PMMA; red, constantan (Ni_{45}Cu_{55}). b. Schematic representation of the device. Current is sent from contact 1 to contact 2, while recording the voltage between contacts 3 and 4. Contacts 1, 2, 5 and 6 are used for four-probe spin-valve measurements. The thermocouple is electrically isolated from the bottom contact by an Al_{2}O_{3} (green) layer.
to compare these values of $I_{ac}$ with the previously reported spin-dependent Seebeck coefficient ($S \propto -S_x$) of $-3.8 \mu V K^{-1}$. Using $I_{ac} = S_x T$ and by taking $T = 300 K$, we obtain values for $S$ between $-3.0 \mu V K^{-1}$ and $-4.3 \mu V K^{-1}$ from our measurements, in agreement with the results of ref. 9.

The observed background signal in the first-harmonic measurement due to the conventional Peltier effect is a factor of 3 lower than we obtained from modelling. We attribute this difference to difficulties in accurately determining the combination of Peltier effects for all the interfaces in the current path. Given these uncertainties, we allow for the possibility that the actual value of the spin-dependent Peltier coefficient may be slightly higher than obtained from the current modelling.

To confirm that the spin-dependent Peltier effect is indeed the origin of the spin signal, the same device was investigated at 77 K (Supplementary Section E). No spin signal could be observed in the $R_{ac}^2$ measurement, as expected from the temperature dependence $I_{ac} \propto T^2$. This dependence is obtained by taking $S(T) \propto T$ in the Thomson–Onsager relation and gives an upper bound of $-0.1 mV$ for $I_{ac}$ from the observed noise level at 77 K.

In conclusion, we have experimentally demonstrated a magnetically controllable heat current, driven by the spin dependency of the Peltier coefficient. The relatively low efficiency of this effect in ferromagnetic metals restricts the cooling or heating power of the device. For use in applications, the effect may be enhanced by the use of non-metallic materials. Spin-dependent Peltier coefficients that are an order of magnitude larger than that in Py will bring the achievable temperature differences to within a range of a few kelvins. With electronic components becoming smaller and smaller, the need for local and programmable refrigeration devices is growing, and it is possible that the spin-dependent Peltier effect might fill this role.

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Author contributions

J.F., F.L.B., A.S. and B.J.v.W. conceived the experiments. J.F. and F.L.B. designed and carried out the main experiments. J.F., F.L.B. and F.K.D. took part in the fabrication process. J.F. and F.L.B. performed the analysis and wrote the paper, with the help of all authors.

Additional information

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