Making flexible spin caloritronic devices with interconnected nanowire networks

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Spin caloritronics has recently emerged from the combination of spintronics and thermoelectricity. Here, we show that flexible, macroscopic spin caloritronic devices based on large-area interconnected magnetic nanowire networks can be used to enable controlled Peltier cooling of macroscopic electronic components with an external magnetic field. We experimentally demonstrate that three-dimensional CoNi/Cu multilayered nanowire networks exhibit an extremely high, magnetically modulated thermoelectric power factor up to 7.5 mW/K²m and large spin-dependent Seebeck and Peltier coefficients of −11.5 μV/K and −3.45 mV at room temperature, respectively. Our investigation reveals the possibility of performing efficient magnetic control of heat flux for thermal management of electronic devices and constitutes a simple and cost-effective pathway for fabrication of large-scale flexible and shapeable thermoelectric coolers exploiting the spin degree of freedom.

INTRODUCTION

The ability of thermoelectric materials to harvest waste heat and deliver cooling power through solid-state devices without moving parts makes them important candidates of sustainable energy technologies in the future. Innovative spin-based transport mechanisms are crucial steps in developing the next generation of thermoelectric materials (1). In this context, coupling heat-driven transport with spintronics is at the heart of the rapidly emerging field of spin caloritronics (2, 3). Previous studies on nanoscale precision magnetic structures such as lithographically defined nanopillars have led to the observation of various spin-enabled mechanisms such as spin Seebeck effects (4, 5), thermally driven spin injection (6), and thermally assisted spin-transfer torque (7, 8). However, key issues to observe the magnetic control of heat flux in a spintronic device are related to insufficient power generation capability and difficulties in detecting extremely small temperature changes in such nanostructures. Although there have been some initial studies on the thermoelectric analogs of giant magnetoresistance (GMR) in magnetic multilayers with current in-plane configuration (9–12), the effects of interfaces make the interpretation of the results more delicate than in the simpler CPP (current-perpendicular-to-plane) configuration (13). In the limit of no-spin relaxation, most of the CPP-GMR data can be understood using a simple two-current series resistor model, in which the resistance of layers and interfaces simply adds and where "up" and "down" charge carriers are propagating independently in two spin channels with large spin asymmetries of the electron’s scattering (14, 15).

Similarly, the spin-dependent thermoelectric effects exploit the fact that the Seebeck coefficients for spin-up and spin-down electrons, $S_\uparrow$ and $S_\downarrow$, are also different because of the exchange splitting of the d-band (16, 17). The 3d ferromagnetic metals exhibit relatively large diffusion thermopowers because of the pronounced structure of the d-band and the high energy derivative of the density of states at the Fermi level. Besides, these magnetic metals also exhibit substantial magnon-dragn contribution to the thermoelectric power within a wide temperature range (18, 19). Moreover, the largest room temperature (RT) power factor (PF = $S^2\sigma$, with $\sigma$ as the electrical conductivity), which is the physical parameter that relates to the output power density of a thermoelectric material, is achieved for cobalt (PF ≈ 15 mW/K²m) (20).

To date, most of the investigations of thermoelectric transport in CPP-GMR systems were performed on lithographically defined nanopillars, single nanowire (NW), and parallel NW arrays (21–24). The thermoelectric properties of nanostructures are more challenging to measure than in bulk materials since it is difficult to determine and/or eliminate contact thermal resistance, an important error source, and simulations are often required to estimate the temperature gradient over the multilayer stacks (6, 23, 25, 26). Therefore, accurate determination of the spin-dependent Seebeck coefficients still remains challenging, and from these previous works, only relatively small RT values of $S_\uparrow$ − $S_\downarrow$ ranging from −1.8 to −4.5 μV/K for cobalt and permalloy, respectively, were indirectly estimated from measurements performed on nanopillar devices (6, 27). In addition, observations of net Peltier cooling in these magnetic nanostructures have not been possible because of the dominant Joule heating effect compared to the Peltier effect together with the technical issues in detecting extremely small temperature changes. Besides, the thermoelectric PFs of CPP-GMR nanopillar and NW systems were not determined from these experiments. Spin-dependent Seebeck and Peltier effects were also recently reported in magnetic tunnel junctions (25, 26, 28). Although magnetic tunnel junctions are expected to be suitable materials for magnetic switching by virtue of thermal spin-torque (8), the high electrical resistance (in the kilohm range) and the nanometer junction size prevent their use for thermoelectric Peltier coolers due to the dominant Joule heating effect. A recent experimental study made on magnetic tunnel junctions leads to change in temperature due to the magneto-Peltier effect of the order of ~100 μK at RT for an electric current density of several kiloamperes per square centimeter (28). So, despite the practical significance, all these experimental issues impose strong restrictions on applications in the burgeoning field of spin caloritronics.

In this study, we developed an experimental platform that allows for the observation of magnetically controlled heat flux in custom-fabricated spin caloritronic devices. Specifically, we fabricated macrosopic interconnected networks made of CoNi/Cu NWs by direct electrodeposition into three-dimensional (3D) nanoporous polymer host membranes (see Materials and Methods) (29, 30). This fabrication strategy allows us to move from submicrometer-scale magnetic nanostructures to more efficient macroscopic-scale spin caloritronics...
devices. Besides, electrical connectivity is essential to allow charge flow over the whole sample sizes. The multilayered NW network films based on the CPP geometry combine numerous advantages, such as the high thermoelectric PF and large magnetothermo-electric effects that can be easily determined using simple but precise measurement setups. Using centimeter-scale NW network films, we demonstrated net Peltier cooling of macroscopic electronic components and efficient control of the heat flow using a magnetic field. Also, our experimental measurements enable reliable determination of key spin-dependent material parameters over a wide temperature range. Another significant advantage of our approach is the achievement of highly efficient flexible and shapeable spin caloritronics devices. Moreover, since there is no sample size limitation, the fabrication method is directly expandable into NW network films with much larger dimensions.

RESULTS
The interconnected nanoporous templates have been prepared by performing a sequential two-step exposure to energetic heavy ions, at angles of +25° and −25°, with respect to the normal of the polycarbonate (PC) membrane surface (for details, see Materials and Methods) (31). Two different 22-μm-thick polymer membranes were used in this study with diameters of 80 and 105 nm and very different porosity characteristics. The spin caloritronic devices are networks of interconnected NWs embedded inside the 3D polymer membranes.

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**Fig. 1. 3D interconnected NW networks.** (A to D) Scanning electron microscopy (SEM) images of self-supported interconnected nanowire (NW) networks with different magnifications, diameters, and packing densities. Low-magnification image showing the 50° tilted view of the macroscopic NW network film with 105-nm diameter and 22% packing density (A) and SEM image at higher magnification showing the NW branched structure (B). Low-magnification image showing the top view of the NW network with 80-nm diameter and 3% packing density (C) and SEM image at higher magnification (D) corresponding to (C). (E) Schematic of the interconnected NW network with alternating magnetic and nonmagnetic layers embedded within the 3D nanoporous PC template.

**Fig. 2. Experimental setups for spin caloritronic measurements.** (A) Device configuration to measure the Seebeck coefficient and the magnetothermoelectric effect. Heat flow is generated by a resistive element, and a thermoelectric voltage $\Delta V$ is created by the temperature difference $\Delta T$ between the two metallic electrodes that is measured by a thermocouple (see Materials and Methods for details). (B) Device architecture and measurement configuration for the Peltier cooling/heating detection. Two Cernox temperature sensors A and B are used to probe the local temperature change at the electrode—NW network junctions induced by the electric current flow (see Materials and Methods for details). Device dimensions in (A) and (B) are 15 mm long, 5 mm wide, and 22 μm thick, and the color represents the generated temperature profile in the NW networks. The gold electrodes are 2 mm wide.
The absolute value of the magnetothermopower $MTP = (\text{field dependencies and relative changes of } \sim 30\%$ at low-packing density CoNi/Cu NW sample show the same magnetic As shown in Fig. 3 (A and B), the resistance and thermopower of the NW networks

Spin-dependent thermoelectric transport in CoNi/Cu NW network films (Fig. 2B) (see Materials and Methods).

The crossed NWs with a multilayer structure of CoNi/Cu were grown in the host porous templates by pulsed electrochemical deposition (32). The thickness of the bilayers was set as 15 nm with approximately the same thickness for the CoNi and Cu layers. Figure 1 (A to D) displays the morphology of self-supported interconnected NW samples obtained after template dissolution, whereas Fig. 1E illustrates the 3D multilayered architecture. As schematically outlined in Fig. 2, we developed two experimental setups allowing to simultaneous measure the magnetic resistance and Seebeck coefficient (Fig. 2A) and to determine the Peltier heating and cooling power generated at the electrodes of the flexible NW network films (Fig. 2B) (see Materials and Methods).

Spin-dependent thermoelectric transport in CoNi/Cu NW networks

As shown in Fig. 3 (A and B), the resistance and thermopower of the low-packing density CoNi/Cu NW sample show the same magnetic field dependencies and relative changes of $\sim 30\%$ at $H = 8$ kOe at RT. The absolute value of the magnetothermopower $MTP = (S_{\text{AP}} - S_p)/S_{\text{AP}}$, with $S_{\text{AP}}$ and $S_p$ as the corresponding thermopowers in the high- and low-resistance states, respectively, shows a similar increase with decreasing temperature as the MR ratio (defined as $MR = (R_{\text{AP}} - R_p)/R_{\text{AP}}$) for temperatures larger than 130 K (see Fig. 3C). However, below this temperature, the MTP exhibits a less pronounced effect and reaches a minimum value around $-25\%$ at $T = 50$ K, where a further increase at lower temperatures results in MTP values up to about $-40\%$.

The Seebeck coefficients at RT are measured to be $-21.3 \mu V/K$ in the AP state and $-27.6 \mu V/K$ in the P state, and the MTP reaches about $30\%$. These values are in agreement with the results obtained from measurements carried out on a single CoNi/Cu NW (24). Calculations of the thermoelectric PF at RT lead to $PF_{\text{AP}} = S_{\text{AP}}^2/\rho_{\text{AP}} \approx 3.1 \text{ mW/K}^2\text{m}$ and $PF_P = S_p^2/\rho_P \approx 7.5 \text{ mW/K}^2\text{m}$ (see section S2 for the estimation of the resistivity values), which are comparable values or even larger than the PF of the widely used thermoelectric material, bismuth telluride (in the range of 1 to 5 mW/K$^2$m) (33). Besides, the PF values obtained for CoNi/Cu NW networks embedded in polymer membranes are at least one order of magnitude larger than those of flexible thermoelectric films based on optimized conducting polymers (34). The magneto-PF ($PF = (PF_{\text{P}} - PF_{\text{AP}})/PF_{\text{AP}}$) reaches $\sim 135\%$ at RT and exceeds $200\%$ at low temperatures, as also shown in Fig. 3D. The efficiency of a material’s thermoelectric energy conversion is determined by its figure of merit $ZT = S^2\sigma T/\kappa$, with $\kappa$ the thermal conductivity. Because of the very low thermal conductivity of PC ($\kappa = 0.2 \text{ W/m K}$ at RT), the contribution of the polymer matrix to heat transport is much smaller than that of the metallic NW network. For metallic NWs, heat transport is dominated by the electron heat conduction flux (35), so assuming that the Wiedemann-Franz law holds, an estimate of the electronic thermal conductivity at RT gives $\kappa_E = 50 \text{ W/m K}$ in the AP configuration and $\kappa_E = 72 \text{ W/m K}$ in the P configuration. In this case, the figure of merit is reduced to $ZT = S^2/L_0$, with $L_0$ the Lorenz number. Using this approximation, we obtain
ZT = 3.1 × 10⁻² at RT at magnetically saturated state for the CoNi/Cu NW sample. Although the RT ZT value in this CoNi/Cu CNW sample is about one order of magnitude smaller than that in BiTe alloys, it is comparable to those of thermocouple alloys (ZT = 6 × 10⁻² and ZT = 1.4 × 10⁻² in constantan and chromel, respectively) and can be used in applications for devices with low energy requirements when the supply of heat essentially is free as with waste heat. Moreover, recent work (36) suggests that traditionally used high-ZT thermoelectric materials are not the more appropriate materials for electronic cooling applications. In case the goal is to cool hot spots, the Peltier current is in the same direction as the natural conduction and materials with larger PFs and large thermal conductivities should be used. In this context, magnetic multilayered NW networks are good candidates for active cooling (36).

Using the parameter \( \beta = (\rho_1 - \rho_f)/(\rho_1 + \rho_f) \) characterizing the asymmetry in the resistivity between the two spin conduction channels in ferromagnetic layers, the spin-dependent Seebeck coefficients \( S_f \) and \( S_c \) can be expressed as follows (see section S3)

\[
S_f = \frac{1}{2} \left[ S_{AP}(1 + \beta^{-1}) + S_P(1 + \beta^{-1}) \right] \quad \text{(1)}
\]

The temperature evolutions of \( S_{AP}, S_p, S_f, \) and \( S_c \) are shown in Fig. 3E. Although magnon-drag may contribute to the thermopower for some temperature range, the Seebeck effect in the NW networks is dominated by the diffusion contribution as indicated by the nearly linear decrease of the Seebeck coefficients with decreasing temperature and the relationship between thermoelectric power and resistivity reported in section S4. The estimated values at RT are \( S_f = -30.2 \mu V/K, S_c = -18.7 \mu V/K, \) and \( S_f - S_c = -11.5 \mu V/K \) using \( \beta = -1.5 \), which are similar to those previously reported in bulk Co \( S_f = -30 \mu V/K \) and \( S_c = -12 \mu V/K \) (17). From Eqs. 1 and 2, it can be easily deduced that \( S_f = S_p \) and \( S_c = S_{AP} \) in the limit of an extremely large MR ratio (\( \beta \rightarrow 1 \)). Since the MTP can also be expressed as MTP = \( 2\beta \eta / (1 + \beta \eta) \), where \( \eta = (S_f - S_c) / (S_f + S_c) \) denotes the spin asymmetry for Seebeck coefficients (see section S3), infinitely large MTP and MPF effects are expected when the product \( \beta \eta \) tends to –1. In previous works performed using both Ni and ferromagnetic dilute alloys with the occurrence of a virtual bound state at the Fermi level (16, 17), \( S_f \) and \( S_c \) were found to have an opposite

\[
S_c = \frac{1}{2} \left[ S_{AP}(1 + \beta^{-1}) - S_P(1 + \beta^{-1}) \right] \quad \text{(2)}
\]

Fig. 4. Direct observation of Peltier, Joule, current crowding, and magneto-Peltier effects. (A) Temperature versus time traces of the sum of the Joule and Peltier heats relative to a working temperature of 320 K, as recorded by the Cernox sensors A and B (see Fig. 2B). A direct current of 2 mA is applied both forward and reverse in the interconnected CoNi/Cu NWs that are 105 nm in diameter and with a packing density of 22% (\( R = 1.8 \) ohms, MR = 6.3%). (B) Same as in (A) but for higher current intensities for which the Peltier effect becomes dominated by the Joule heating, i.e., \( I = 20 \) and 40 mA, and restricted to data recorded at sensor B. Both in (A) and (B), the DC current is switched on after 100 s as shown by the vertical dashed lines. (C) Measured temperature changes \( \Delta T_f \) at the Peltier junction B during the magnetic field sweep for DC currents of –30 and +30 mA. Here, the contribution from the Peltier heating has been estimated (section S5). The Peltier term leads to heating and cooling at the saturation field of 9.5 kOe and depends on current flow direction. (D) Measured total temperature changes \( \Delta T_{net} \) at the Peltier junction B between the zero-field \( (T_{net}) \) and saturated states \( (T_{net}) \) versus current intensity applied both forward and reverse. Inset: Data obtained in the low-current range. The error bars in (C) and (D) reflect the uncertainty of the temperature measurements as described in section S6.
sign, which corresponds to $|\eta| > 1$. Therefore, the fabrication of multilayered NWs with appropriate magnetic layer composition should make it possible to fine-tune the PF of thermoelectric energy conversion with an external magnetic field.

**Magnetic control of Peltier cooling**

Current flow in the high-packing density CoNi/Cu NW sample results in Joule heating and a Peltier heat current at the junctions between the NW network and the gold electrodes. These heat flows were monitored continuously from the temperature changes using Cernox sensors (A and B, as shown in Fig. 2B) with respect to the operating temperature $T = 320$ K for different current intensities and polarities, as well as for various applied magnetic fields. Net cooling occurs at low currents when the direction of the Peltier heat current flow is such that $\Pi I < 0$ at the considered junction (with $\Pi$ as the Peltier coefficient) and dominates over the Joule heating effect ($R^2 I^2$), as shown in Fig. 4A. When the DC current flows from the NWs (with the higher Peltier coefficient) to the gold electrode (with the lower one), the Peltier heat is released from the junction, i.e., net cooling happens for $I = +2$ mA at sensor B, while net heating happens at sensor A (see Fig. 4A). As expected, the situation is reversed when the current flows in the opposite direction. For currents larger than 5 mA, the Joule heating dominates over the Peltier cooling, as shown by two representative temperature versus time traces for $I = \pm 20$ and $\pm 40$ mA (see Fig. 4B). Unlike the Peltier effect that leads to a cooling or a heating of the system depending on the direction of current flow, the Joule effect does not depend on the current polarity. It is thus possible to separate the two effects linearly (see section S5). From Fig. 4B, one can estimate the Peltier cooling ability of the NW network of $\approx 7.5$ K/A.

As shown in Fig. 4C, the magneto-Peltier effect has been quantified by recording the temperature change $\Delta T_{\text{TH}}$ during the magnetic field sweeps. The field dependence of $\Delta T_{\text{TH}}$ resembles that of MR. The same measurements were performed for different currents, as shown in Fig. 4D, where the total temperature change ($\Delta T_{\text{TH tot}} = T_{\text{TH}} - T_{\text{TH sat}}$) between the zero-field ($T_{\text{TH}}$) and saturation ($T_{\text{TH sat}}$) states is reported. As expected, the magneto-Peltier effect increases linearly with the driving current. From these results, one can obtain the guideline for the magnitude of the magnetically controlled cooling and heating ability of a macroscopic electronic component against the injected current. Using the values of $S^\uparrow$ and $S^\downarrow$ from Fig. 3E and the Onsager relation, which relates the two thermoelectric coefficients $\Pi = ST$, one may estimate the maximum difference between the Peltier coefficients in the AP and P states for an infinite MR ratio as $\Pi^\uparrow - \Pi^\downarrow$, which correspond to $-3.45$ mV at RT for CoNi/Cu NWs. Since currents up to a few hundreds of milliamperes are able to pass through the densely packed NW films without damaging the network structure, we may anticipate that a magnetic field can switch a heat flow as large as 1 mW.

**DISCUSSION**

We have experimentally demonstrated magnetic field control of the Peltier cooling of electronic components using efficient and macroscopic-scale spin caloritronic devices built from CoNi/Cu NW networks. We have found very high thermoelectric PF up to 7.5 mW/K²m at RT, which are larger values than the PF of the widely used thermoelectric material, bismuth telluride. The PF is magnetically modulated with a PF change ratio greater than 150%.

Moreover, key spin-dependent material parameters were extracted accurately over a wide temperature range. Large spin-dependent Seebeck and Peltier coefficients of $-11.5$ and $-3.45$ mV were obtained at RT, respectively.

This work should stimulate further systematic exploration and optimized growth of a variety of 3D NW networks with high magnetothermoelectric performance. We anticipate that the use of less-resistive NW network films with larger packing factor and thickness up to 100 $\mu$m should significantly enhance the ability of net refrigerating effect controlled by a magnetic field. The dimensions of such 3D NW networks are highly scalable, and we can anticipate that they might be used as shapeable thermoelectric components for hot-spot cooling of electronic devices. In principle, realization of planar NW-based thermoelectric modules as small as $\sim 1$ mm² can be made. Such dimensions are appropriate to fit a variety of practical applications requiring effective temperature management of sensors and integrated circuits. In addition, a practical thermoelectric cooler made of flexible and lightweight thermoelectric modules consisting of stacked NW network films that are connected electrically in series and thermally in parallel can be obtained. These observations hold promise for active cooling of electronic devices and waste energy harvesting using extremely light and flexible thermoelectric generators and could lead to advances in future spin caloritronic devices.

**MATERIALS AND METHODS**

**Sample fabrication**

The PC porous membranes with interconnected pores have been fabricated by exposing a 22-μm-thick PC film to a two-step irradiation process. The topology of the membranes was defined by exposing the film to a first irradiation step at two fixed angles of $-25^\circ$ and $+25^\circ$ with respect to the normal axis of the film plane. After rotating the PC film in the plane by $90^\circ$, the second irradiation step took place at the same fixed angular irradiation flux to finally form a 3D nanochannel network. The diameter of the latent tracks was enlarged by following a previously reported protocol to obtain membranes with distinct porosities and pores sizes (37). The PC membranes with average pore diameters of 80 and 105 nm display low volumetric porosity (3%) and large volumetric porosity (22%), respectively. Next, the PC templates were coated on one side using an e-beam evaporator with a metallic Cr/Au bilayer to serve as cathode during the electrochemical deposition. The thickness of the thin adhesion layer of Cr was 3 nm, while for a uniform and consistent nanopore coverage withstanding the electrodeposition process, the Au film thickness was set to 400 and 750 nm for the 80- and 105-nm-diameter porous membranes, respectively.

The multilayered NW networks have been grown at RT by electrodeposition into the 3D porous PC templates from a single sulfate bath using potentiostatic control and a pulsed electrodeposition technique (38). For these experiments, we used an Ag/AgCl reference electrode and a Pt counter electrode. To prepare the CoNi/Cu interconnected NW networks, the composition of the electrolyte was 2.3 M NiSO₄·6H₂O + 0.4 M CoSO₄·7H₂O + 15 mM CuSO₄·5H₂O + 0.5 M H₂BO₃, and the deposition potential was alternatively switched between $-1$ V to deposit equiatomic CoNi alloy layer (containing approximately 5% Cu impurity), and $-0.4$ V to deposit almost pure Cu layers (39). Following a procedure described elsewhere (38), the deposition rates of each metals were determined from the pore filling time. According to this calibration, the deposition time was adjusted...
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SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/5/3/eaav2782/DC1

Section 51. Basic magnetic, magnetoresistance, and thermoelectric property characterization

Section 52. Estimation of the resistivity for the CoNi/Cu NW network

Section 53. Expression for the diffusion thermopower in the two-current model

Section 54. Relationship between the field-dependent thermopower and electrical resistance

Section 55. Data on magneto-Peltier

Section 56. Experimental measurement uncertainty evaluation

Fig. 51. Magnetic characterization curves.

Fig. 52. Gorter-Nordheim characteristics.

Fig. 53. Peltier temperature measurements at the Peltier junction B

Fig. 54. Magneto-Joule and magneto-Peltier temperature measurements at the Peltier junction B

References (40–42)
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