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Magnetothermal effects in lateral ferromagnet-normal metal interfaces:

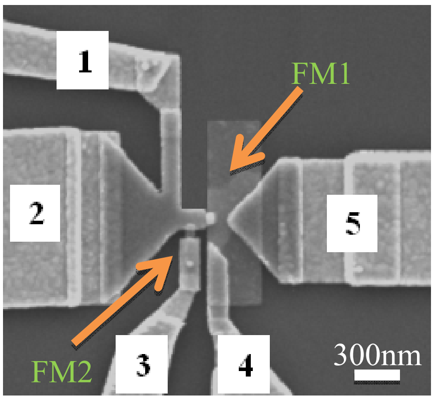
Spin dependent Seebeck effect ; spin dependent Peltier effect and anomalous-Nernst effects

1. Introduction

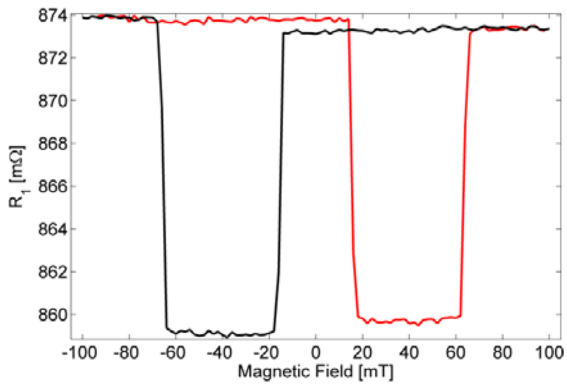
In metallic nanostructures, interactions between different forms of current (charge, spin or heat) result in numerous remarkable effects. Spin caloritronics studies the interaction between charge, spin and thermoelectric effects in systems consisting of a normal metal (NM) and a ferromagnet (FM). From a collection of thermal and thermomagnetic effects: Seebeck, Peltier, Nernst-Ettingshausen, and Righi–Leduc Effect, to mention a few, here we report on our recent works on the determination of spin Seebeck coefficient of $Ni_{80}Fe_{20}$ and Cobalt, observation of spin Peltier effect and the anomalous-Nernst effect in $Ni_{80}Fe_{20}$/Cu and Co/Cu lateral and pillar spin valve systems.

1 Spin dependent Seebeck effect or thermally driven spin injection

When a charge/heat current flows through a FM/NM interface spins accumulate at the interface due to abrupt changes in conductivity/Seebeck coefficients of spin-up and spin-down electrons. This is termed as electrical/thermal spin injection. Basically, electrical spin injection and thermal spin injection are related to each other and can well be described by spin dependent Boltzmann theory. We prepared $Ni_{80}Fe_{20}$/Cu non-local spin valve devices [Figure-1a] to validate thermal spin injection at the $Ni_{80}Fe_{20}$/Cu interface and compare the spin Seebeck coefficient of $Ni_{80}Fe_{20}$ with results obtained in a different device geometry [1]. Charge current j from contact 1 to 5 injects a spin current $j_s = j_\uparrow - j_\downarrow$ creating non-equilibrium spin accumulation at the $Ni_{80}Fe_{20}$/Cu interface that can be probed by measuring the voltage with contacts 2 and 3. The measured non-local spin valve signal ranged from $10\text{-}14 m\Omega$ for the new device geometries.



A)



B)

Figure 1: A) Non-local spin caloritronic device used to measure thermal spin injection. Two $Ni_{80}Fe_{20}$ ferromagnets, FM1 and FM2 are connected by a 60-nm-thick copper funnel with small effective FM/NM contact area of 40×40nm2 for efficient spin injection. Thick Ti/Au contacts 4 and 5 are used to Joule heat FM1 while contacts 2 and 3 are used to measure electrical/thermal spin injection. B) The spin valve effect in a non-local spin valve.

In a similar manner, heat current flowing at the $Ni_{80}Fe_{20}$/Copper interface creates a spin current that flows into the NM. A thermal gradient at the $Ni_{80}Fe_{20}$/Cu was created by sending charge current from contact 4 to contact 5 thereby Joule-heating the ferromagnet locally. Heat current $Q = -\kappa \nabla T$ through the $Ni_{80}Fe_{20}$/Cu interface creates a spin accumulation $\mu_s = \mu_\uparrow - \mu_\downarrow \propto \lambda_F S_s \nabla T \propto j^2$ that flows into the NM. Here $S_s = S_\uparrow - S_\downarrow$ is the spin Seebeck coefficient of the ferromagnet, P_s is the polarization of the Seebeck coefficient and ∇T is the applied thermal gradient. It is inevitable that small percentage of the charge current also flows through the small $Ni_{80}Fe_{20}$/Cu interface resulting in regular spin injection. Hence the signal we measure is non-linear:

$$V = R_1 I + R_2 I^2 + R_3 I^3 \ldots \qquad [1]$$

By using a standard lock-in measurement, we identified the contributions of the linear and non-linear (Seebeck) effects in the signal. For each measurement the signal is fed to three different lock-in amplifiers which are set to measure the specific harmonics of the signal. The spin dependent Seebeck effect being linear on the thermal gradient ($\propto j^2$) shows up in the 2^{nd} harmonic while the Peltier effect shows up in the 1^{st} harmonic. Results of thermal spin injection measurements for newly designed $Ni_{80}Fe_{20}$/Cu [Figure 2a] and Co/Cu [Figure 2b] devices are shown below. For 2mA of current the 2^{nd} harmonic signal due to Joule heating of FM1 is $-51.92nV$ for $Ni_{80}Fe_{20}$/Cu and $-101.3nV$ for Co/Cu samples. The coercive fields of the Cobalt ferromagnets are also observed to be higher than that of $Ni_{80}Fe_{20}$.

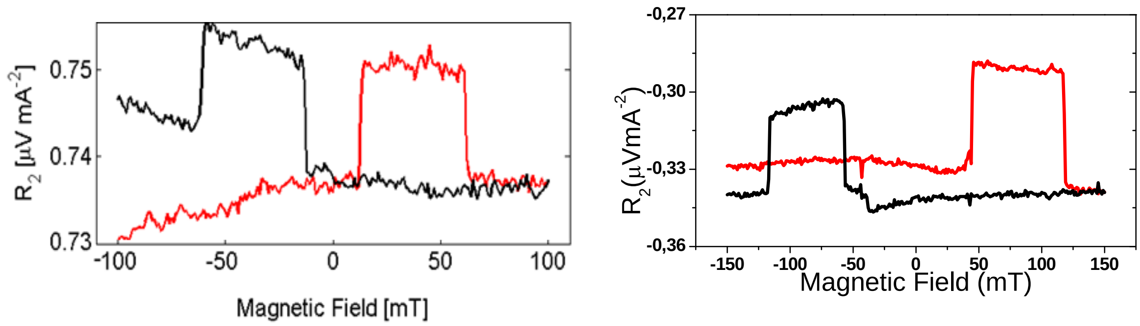


Figure 2: Second Harmonic signal measured for A) $Ni_{80}Fe_{20}$/Cu and B) Co/Cu nonlocal spin valve devices. The baseline resistance, originating from Joule heating, is higher in A than in B due to the low resistivity of Cobalt.

The efficiency of thermal spin injection solely depends on the bulk spin polarization, Seebeck coefficient and spin relaxation length of the ferromagnet. Comparative study of thermal spin injection in $Ni_{80}Fe_{20}$/Cu, Co/Cu and Ni/Cu systems shows that indeed the efficiency of the thermal spin injection can be improved with an optimum device design and combination of materials. An attempt to observe thermal spin injection in Ni/Cu was unsuccessful as it was not even possible to achieve conventional spin injection, probably for reasons mentioned in [Jedema et al.,PRB, (2003)]. Nevertheless, from the measurement on 4 samples it is clearly visible that the signals are indeed as expected [cf. Figure 3a and 3b]. However, due to sticking problem of Cobalt on SiO_2 and its heavy oxidation before the next lithography step, it was not possible to confirm this result in other batches of samples.

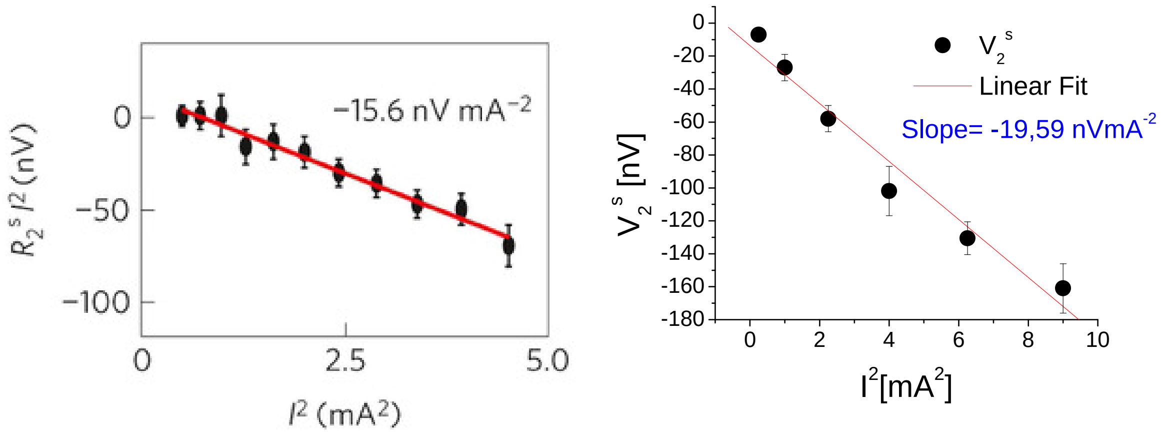


Figure 3: Measured 2nd harmonic voltage V_2(nV) as a function of I^2 for A)) $Ni_{80}Fe_{20}$ /Cu and B) Co/Cu non local spin valves.

To analyze the data further we constructed a 3D spin-thermoelectric model of the devices using the finite-element program Comsol Multiphysics. By using Mott's two-current model we defined the physics in terms of a thermoelectric spin model where the spin-up, down and heat currents are given by:

$$\begin{pmatrix} J_\uparrow \\ J_\downarrow \\ Q \end{pmatrix} = -\begin{pmatrix} \sigma_\uparrow & 0 & \sigma_\uparrow S_\uparrow \\ 0 & \sigma_\downarrow & \sigma_\downarrow S_\downarrow \\ \sigma_\uparrow \Pi_\uparrow & \sigma_\downarrow \Pi_\downarrow & \kappa \end{pmatrix} \begin{pmatrix} \nabla V_\uparrow \\ \nabla V_\downarrow \\ \nabla T \end{pmatrix} \quad [2]$$

where $\Pi_{\uparrow,\downarrow}$ are the spin-dependent Peltier coefficients given by $S_{\uparrow,\downarrow} \cdot T_0$. Here T0=300K, which is the reference temperature of the device. Yet another promising direction for this experiment is to perform similar experiments in a pillar structure where the interface polarization is higher due to in-situ fabrication of the device structure. A typical device structure of this type is shown in Figure 4 where a stack of $Ni_{80}Fe_{20}$/Cu/ $Ni_{80}Fe_{20}$ is sandwiched between a Pt bottom contact and Gold top contact. In such a way it is possible to do a comparative study of thermal spin injection experiment in different FM-NM combinations.

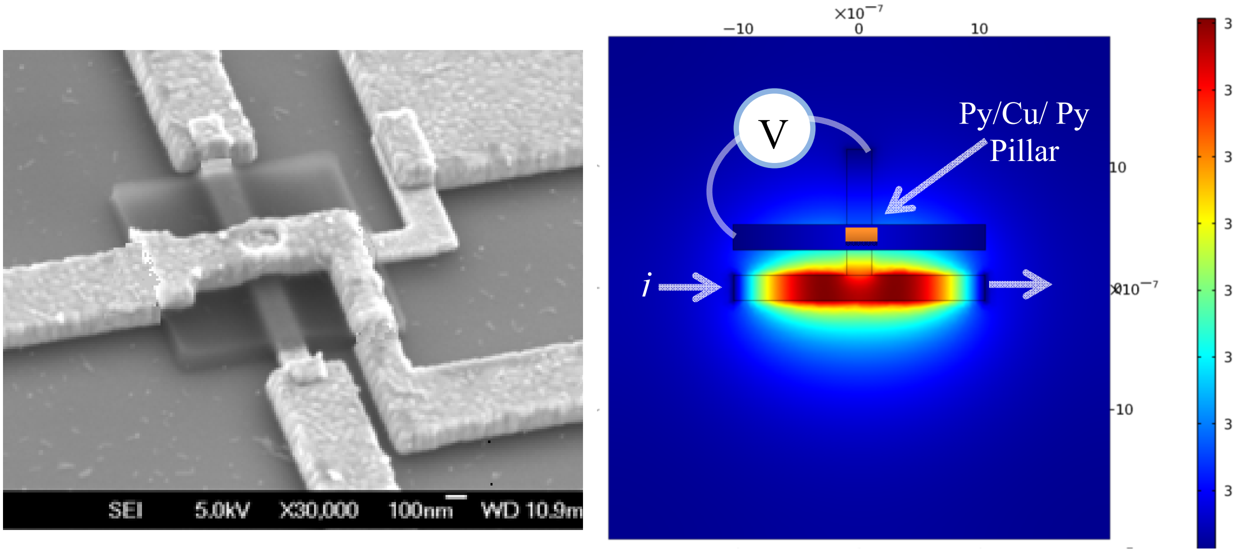


Figure 4: Pillar structures prepared for measuring thermal spin injection for $Ni_{80}Fe_{20}$/Cu/$Ni_{80}Fe_{20}$, Co/Cu/Co, systems. A stack of $Ni_{80}Fe_{20}$/Cu/ $Ni_{80}Fe_{20}$ is sandwiched between Platinum bottom contact and Gold top contact. Cross-linked PMMA is used to electrically isolated top and bottom contacts. B) The scheme used to create the thermal gradient. Heat created by Joule- heating the Pt bottom contact diffuses to the left and heats up the bottom FM contact. Consequently a thermal gradient is created in the z-direction that create by sending charge current at the bottom contact heat diffuses away through the Platinum arm where the pillar is deposited on.

2 Anomalous-Nernst (AN) effect and magnetoresistive heating

The AN effect is the thermal analogue of anomalous-Hall Effect where a transverse voltage gradient perpendicular to both the magnetization and the heat current directions is generated (figure 3). The AMR effect is the dependence of the resistivity of the ferromagnet on the angle between the current flow and its magnetization. Both effects can be modeled by taking into account the anisotropy in σ and the Seebeck coefficient S as:

$$\sigma_{ij} = \sigma_{\perp}\left(\delta_{ij} - R_{AMR} m_i m_j\right)$$
$$S_{ij} = S\left(\delta_{ij} - R_N \sum_k \varepsilon_{ijk} m_k\right) \qquad [2]$$

In our thermal spin injection experiments, it is inevitable that the detecting ferromagnet picks up anomalous-Nernst signals on top of the thermal spin injection signal. Joule heating of FM1 induces a heat flow $Q = -k\nabla T\hat{x}$ through FM2 (magnetization along $\hat{y}$) which creates a transverse voltage given by $\vec{\nabla} V_N = -S_N \vec{m} \times \vec{\nabla} T$ along the $\hat{z}$ direction. Naturally, the heat arriving at FM2 depends on the resistivity of FM1 which depends on the angle between the direction of current and magnetization of FM1.We performed study of these two effect in a dedicated spin caloritronic device—the results of which is published in PRB as rapid communication. For a detailed study of anomalous-Nernst and anisotropic magnetoresistive heating in a caloritronic device refer to [4].

3 Spin Peltier effect

The observation of spin Seebeck effect and the realization of thermally driven spin injection across a FN/NM interface[1] poses a natural question as to how one can observe the spin Peltier effect. Since the spin Peltier effect is related to the spin Seebeck effect by $\Pi = ST_0$, where Π is the Peltier coefficient, S is the Seebeck coefficient and T_0 is the reference temperature; it should in principle be possible to measure this effect. This effect can occur when there is pure spin current arriving at the 2nd ferromagnet. However, it is also possible that a combination of charge and spin current can arrive at FM2 where in this case a complete analysis is required to identify contributions from conventional Peltier and spin Peltier.

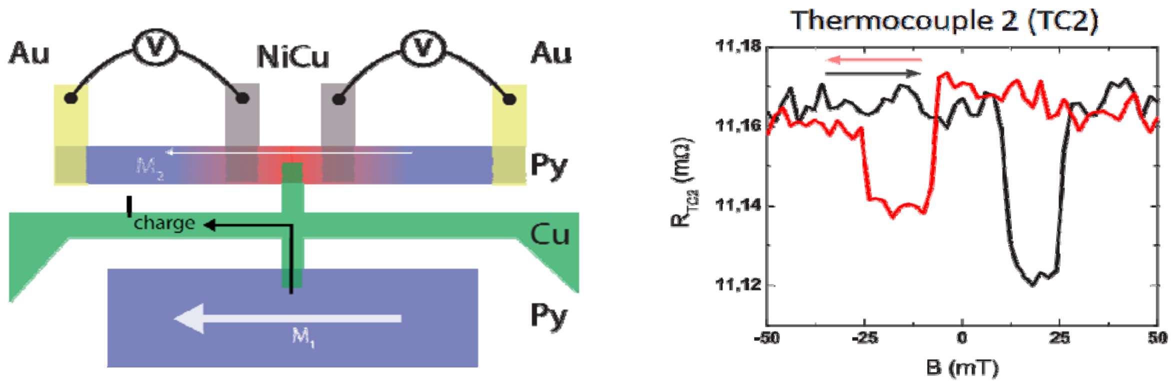


Figure 5: Detailed schematic of how we can measure the spin Peltier effect. A) Device geometry employed to measure spin Peltier effect in NLSV. Spins injected from Py1 into a Cu cross (green) diffuse and enter Py2 resulting in either heating or cooling of the interface depending on the magnetization of Py2. Thermocouples of NiCu/Py detect the local resulting temperature relative to a reference temperature. B) Typical measurement where a clear spin dependent temperature profile is measured by the thermocouple as the function of the applied external magnetic field.

A typical geometry of our devices looks like the one shown in Fig. 5a. We design our devices such that we exclude the regular spin valve signal that can potentially overrule the spin Peltier signal unless optimal design is used. With first batches of samples (Fig. 1a) we measured the spin Peltier effect as shown in Fig. 1b. A spin current is injected into copper by sending charge current through FM1/NM interface. The spin current diffuses and enters FM2 thereby heating/cooling the interface depending on the magnetization direction of the magnetization. To measure the temperature we put highly sensitive thermocouples ($-40\,\mu V/K$) nearby the interface. A result of such measurement is shown in Figure 5b. The voltage measured at the thermocouples shows clear dependence on the relative magnetization orientation of the two ferromagnets. From 3D finite element modelling it was possible to see that a regular non-local spin valve signal ~20μΩ on top of a spin Peltier signal~18μΩ is measured. Another device design shown in Figure 6a was also measured. In this device design we expect a spin Peltier signal of ~22μΩ on top of ~3μΩ spin valve signal. A clear indication to the observation of this effect was to measure at low temperatures. We expected the spin dependent Peltier signal to diminish and eventually vanish while cooling the system down. However, we actually observed enhancement of the signal with decrease in temperature which confirms that we still pick-up part of the spin valve signal. To tackle this problem we can place the thermocouple even further away at the cost of reducing the sensitivity of the thermocouples or increase the spin current arriving at the second ferromagnet by using a pillar spin valve structure instead of a nonlocal lateral spin valve.

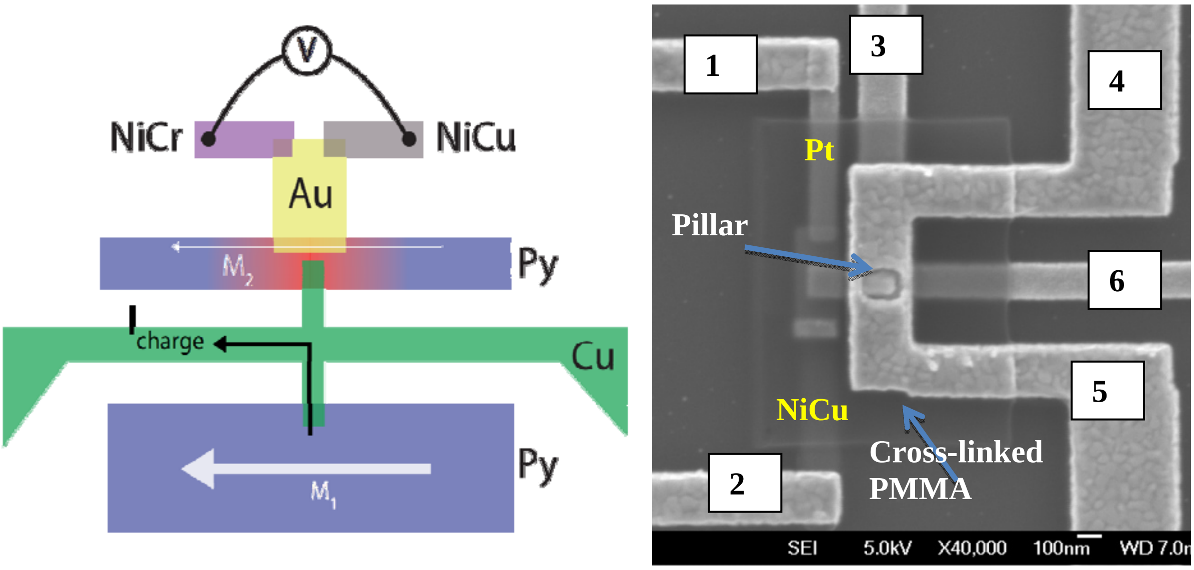


Figure 6: a) Batch II-nonlocal spin valves prepared for spin Peltier effect B) Batch III- device layout used to demonstrate spin Peltier effect. A pillar of $Ni_{80}Fe_{20}/Cu/Ni_{80}Fe_{20}$ is sandwiched between a Pt bottom contact and Au top contact. Cross-linked PMMA is used to isolate the top contact from the bottom contact.

Figure-6b shows the last measured sample to demonstrate the spin Peltier effect. Here an electrically isolated and highly sensitive thermocouple made of platinum and NiCu (contacts 1 and 2) were placed nearby a $Ni_{80}Fe_{20}$/Cu/ $Ni_{80}Fe_{20}$ stack. The bottom Pt contacts (contacts 3 and 6) are isolated from the thermocouples by 8nm of Al_2O_3 such that no spurious signals are picked up by the thermocouples. When a charge current is sent from contact 3 to 4, it creates spin and charge current in the copper spatial layer which diffuses to the bottom FM. While entering the FM it either heats/cools the FM depending on its magnetization direction. Heat created this way diffuses to where the thermocouples are placed. Since the thermocouple is isolated electrically by an Al_2O_3 barrier, the voltage measured at contacts 1 and 2 is due only to the heat. In this case the heat arriving at the thermocouple has to contribution –regular Peltier and spin Peltier effect. By sweeping the magnetic field it is observed that the signal is field dependent confirming the presence of spin Peltier effect. By doing the measurement at low temperatures it was also possible to see that the signal vanishes at 77K confirming the presence of the spin Peltier effect. We still have to further confirm this by replacing the thermocouples from Pt/Nicu to Pt/Pt. In this case the thermocouples should measure zero if the effect is due to heat. We expect to submit this result for publication soon.

2. Summary & Plan

We have performed thermal spin injection experiments on $Ni_{80}Fe_{20}$/Cu and Co/Cu systems and validated the observation of thermally driven spin injection in ref. [1]. We developed a dedicated 3D finite element modeling based on spin dependent Boltzmann theory to explain the results obtained from measurements [1 - 5]. With this model it was also possible to explain spin-orbit coupling related thermal effects as shown by Slachter et al. [4]. As part of the deliverables for the next year we plan to perform a comparative study of spin dependent Seebeck effect and spin Peltier effect for different FM/NM combination. Specifically, a comparative study of spin dependent Seebeck and Peltier effect will be performed for Co/Cu, Ni/Cu, $Co_{84}Fe_{16}$/Cu and NiMnSb/Cu systems after which a publication is expected before the next meeting in September 2012.

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