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Thermal creation of a spin current by Seebeck spin tunneling

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The thermoelectric analog of spin-polarized tunneling, namely Seebeck spin tunneling, is a recently discovered phenomenon that arises from the spin-dependent Seebeck coefficient of a magnetic tunnel contact. In a tunnel junction with one ferromagnetic electrode and one non-magnetic electrode, a temperature difference between the two electrodes creates a spin current across the contact. Here, the basic principle and the observation of Seebeck spin tunneling are described. It is shown how it can be used to create a spin accumulation in silicon driven by a heat flow across a magnetic tunnel contact, without a charge tunnel current. The sign of the spin current depends on the direction of the heat flow, whereas its magnitude is anisotropic, i.e., dependent on the absolute orientation of the magnetization of the ferromagnet. The connection between Seebeck spin tunneling and the tunnel magneto-Seebeck effect, observed in metal magnetic tunnel junctions, is also clarified. Seebeck spin tunneling may be used to convert waste heat into useful thermal spin currents that aid or replace electrical spin current, and thereby improve the energy efficiency of spintronic devices and technologies.

Keywords: Spintronics, Seebeck spin tunneling, spin current, silicon

1. INTRODUCTION

Thermoelectric systems enable the conversion of heat flow into electrical power, and vice-versa. They are based on the interplay between heat and charge transport within materials, and across boundaries between different materials. Optimizing the efficiency of thermoelectric systems is an important goal in the quest for energy saving. Low power consumption and energy efficiency are also important driving factors for the field of spintronics,¹⁻⁴ in which the spin of electrons is the state variable that represents digital information. It has transformed magnetic data storage technology and is expected to do the same for computing technology.¹⁻⁴ Thermoelectrics and spintronics have for the most part developed independently, although the connection between them has been established in studies performed during the last three decades on the coupling between heat and spin transport in nanostructures containing ferromagnetic materials.⁵⁻¹⁵ The interplay between heat and spin transport, now referred to as spin caloritronics,^{14,16,17} has recently gained impetus because it was realized that the combination of thermoelectrics and spintronics offers unique possibilities for the development of green technologies. On the one hand, it provides a new, spin-based approach to thermoelectric power generation and cooling. On the other hand, it provides a thermal route to create and control the flow of spin in novel spintronic devices that make functional use of heat and temperature gradients. In addition, most spintronic nanodevices involve the application of electrical currents, which unintentionally create thermal gradients that influence magnetic and spin-related phenomena and thereby device performance. This underpins the importance of understanding the interactions between thermal and spin effects and motivates the recent push in the field of spin caloritronics.

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Spin caloritronics has recently been reviewed.¹⁷ Here, we limit ourselves to phenomena that derive explicitly from the spin dependence of thermoelectric effects, such as the Seebeck effect, in ferromagnets and their junctions. Electrons with majority ($\uparrow$) and minority ($\downarrow$) spin occupy states with different character, density and energy dispersion. Hence, when a ferromagnetic conductor or junction is subjected to a thermal gradient ∇T , different charge currents $I^\uparrow$ and $I^\downarrow$ are induced for majority and minority spin. Consequently, there is a net flow of spin along the direction of the thermal gradient. Here we shall focus on Seebeck spin tunneling. This pure interface effect occurs in tunneling and was first observed by Le Breton et al.¹⁸ They created a temperature difference across a magnetic tunnel contact to a semiconductor and observed the induced flow of spin-angular momentum across the tunnel barrier. Effectively, it was demonstrated that $S^\uparrow \neq S^\downarrow$, i.e., that the Seebeck coefficient of a ferromagnetic tunnel interface depends on spin. This was first explicitly stated in subsequent theory work.^{19,20}

2. SPIN-DEPENDENT SEEBECK COEFFICIENT OF A TUNNEL JUNCTION

Seebeck spin tunneling can be described within a recently developed phenomenological framework.¹⁹ Consider a tunnel junction with a ferromagnetic electrode and a semiconductor (Fig. 1, left panel), with tunnel resistance R_{tun} . We define $\Delta T = T_n - T_{fm}$, where T_n and T_{fm} are the temperatures of the non-magnetic and ferromagnetic electrode, respectively, and $V = V_n - V_{fm}$, where V_n and V_{fm} are the spin-averaged electrochemical potentials of the non-magnetic and the ferromagnetic electrode. For a given V and ΔT , the tunnel current for each spin is:

$$I^\uparrow = G^\uparrow \left(V - \frac{\Delta\mu}{2} \right) + L^\uparrow \Delta T \quad (1)$$

$$I^\downarrow = G^\downarrow \left(V + \frac{\Delta\mu}{2} \right) + L^\downarrow \Delta T \quad (2)$$

The first term on the right-hand side describes the electrically driven current, taking into account the shifts of the electrochemical potentials in the semiconductor due to the induced spin accumulation $\Delta\mu = \mu^\uparrow - \mu^\downarrow$. The last term describes the thermally-induced tunnel current, with the spin-dependent coefficients $L^\uparrow$ and $L^\downarrow$. These are related to the spin-dependent Seebeck coefficients of the tunnel contact via $S^\uparrow = -L^\uparrow/G^\uparrow$ and $S^\downarrow = -L^\downarrow/G^\downarrow$, where $G^\uparrow$ and $G^\downarrow$ represent the tunnel conductance for majority and minority spin, respectively. At zero charge tunnel current ($I = 0$), the thermally-induced spin accumulation in the semiconductor is given by:¹⁹

$$\Delta\mu = \left\{ \frac{(1 - P_G^2) r_s}{R_{tun} + (1 - P_G^2) r_s} \right\} (S^\uparrow - S^\downarrow) \Delta T \quad (3)$$

where P_G is the tunnel spin polarization and r_s the so-called spin resistance of the semiconductor.²¹⁻²³ Importantly, the thermally-induced spin accumulation is proportional to $(S^\uparrow - S^\downarrow) \Delta T$.

Figure 1. (Left) Energy band diagram of a ferromagnet/insulator/semiconductor junction. The semiconductor is at temperature T_n , the ferromagnet is at T_{fm} . A spin splitting $\Delta\mu = \mu^\uparrow - \mu^\downarrow$ of the electrochemical potential exists in the semiconductor. (Right) Schematic illustration of the thermally-driven spin injection across a tunnel barrier.

The thermal spin accumulation can be probed electrically because, just as the charge thermovoltage ($S \Delta T$, with the charge Seebeck coefficient S), the spin accumulation contributes to the total voltage across the junction:

$$V = R_{tun} I + P_G \left(\frac{\Delta \mu}{2} \right) + S \Delta T \quad (4)$$

The thermal spin current across a tunnel contact is determined by the spin-dependent Seebeck coefficient of the tunnel interface (Fig. 1, right panel). The thermal spin current (and temperature gradient) within the ferromagnet itself is not relevant if the tunnel barrier dominates the transport.^{19,20} The thermal spin current in the ferromagnet will adjust itself near the tunnel interface by inducing a spin accumulation in the ferromagnet. The thermal spin current in the ferromagnet is important for thermally-driven spin injection across metallic interfaces,²⁴ which can be well described in terms of the difference of the Seebeck coefficient of majority and minority spin electrons *in* the ferromagnet.^{24,25} Thus, in metallic contacts the thermal spin current originates from the bulk of the ferromagnet, whereas in Seebeck spin tunneling it is generated by the tunnel interface itself.

3. SEEBECK SPIN TUNNELING AND TUNNEL MAGNETO-SEEBECK EFFECT

In order to clarify the relation between Seebeck spin tunneling and other spin-dependent thermoelectric phenomena in tunnel contacts, such as the tunnel magneto-Seebeck effect²⁶ (also known as the tunnel magnetothermopower²⁷), we make the analogy with electrical spin tunneling (Fig. 2). Since the pioneering experiments by Tedrow and Meservey^{28,29} in the early 1970's on ferromagnet/insulator/superconductor tunnel junctions, it is well known that the electrical tunnel conductance depends on spin ($G^\uparrow \neq G^\downarrow$). This phenomenon, termed spin-polarized tunneling, was later employed in magnetic tunnel junctions with two ferromagnetic electrodes, in which a large change in tunnel resistance (so-called tunnel magnetoresistance (TMR)) is produced when the relative direction of magnetization of the two electrodes is changed.^{30–35} Importantly, spin-polarized tunneling also creates a spin current across the tunnel barrier. This is widely used to exert a torque on the magnetization, or to electrically inject spin currents into non-magnetic materials,^{1–3} including semiconductors such as silicon.⁴ Analogously, Seebeck spin tunneling not only produces thermal spin currents in contacts with one ferromagnetic electrode,¹⁸ but it also underlies the tunnel magneto-Seebeck effect,^{26,27} i.e., a dependence of the thermopower of a magnetic tunnel junction on the relative magnetization alignment of the two ferromagnetic electrodes.

Figure 2. Analogy between spin-polarized tunneling (spin-dependent conductance $G^\uparrow \neq G^\downarrow$) and Seebeck spin tunneling (spin-dependent Seebeck coefficient $S^\uparrow \neq S^\downarrow$). The former underlies tunnel magnetoresistance (left) and electrical spin injection (right) in junctions with, respectively, two or one ferromagnetic electrode. Similarly, Seebeck spin tunneling is the fundamental phenomenon that governs thermal spin injection by tunneling in junctions with one ferromagnetic electrode, but also the tunnel magnetothermopower (or tunnel magneto-Seebeck effect) in junctions with two ferromagnets.

The analogy can be extended. In spin-polarized tunneling, a so-called tunnel spin polarization P can be associated with an interface between a ferromagnet and an insulator. In a tunnel junction with one ferromagnetic and one non-magnetic electrode, the spin current is proportional to P , whereas in a junction with two ferromagnets, the TMR can be expressed in terms of the product $P_1 P_2$ of the tunnel spin polarizations of the first and second ferromagnet/insulator interface. While this simple description breaks down for coherent tunneling, it works well for tunneling in the presence of sufficient disorder, for instance with amorphous tunnel barriers.³⁶ The thermal spin current induced by Seebeck spin tunneling can be expressed¹⁹ in terms of $S^\uparrow - S^\downarrow$, which is proportional to the spin polarization P_S of the Seebeck coefficient. It is plausible, at least for incoherent tunneling, to express the tunnel magnetothermopower^{26,27} in junctions with two ferromagnetic electrodes in terms of the product $P_{S,1} P_{S,2}$ of the Seebeck spin polarization of the first and second ferromagnet/insulator interface.

4. EXPERIMENTAL OBSERVATION OF SEEBECK SPIN TUNNELING

In order to observe Seebeck spin tunneling, we take a ferromagnet/insulator/semiconductor tunnel junction and apply a heating current with density J_{heating} to one of the electrodes (silicon in Fig. 3). This causes Joule dissipation in the Si and raises the temperature of the Si with respect to that of the ferromagnetic electrode. The temperature difference ΔT drives a spin current into the Si and induces a spin accumulation $\Delta\mu$ in the silicon. The thermally-induced spin accumulation is probed by setting the charge tunnel current to zero and measuring the change in voltage in response to an applied magnetic field B in the Hanle geometry, with B along the z-axis perpendicular to the in-plane magnetization of the ferromagnet and to the injected spins. This causes spin precession and a reduction of $\Delta\mu$ to zero with a characteristic Lorentzian line shape.^{37–39} The voltage across the tunnel contact is then reduced by $P_G \Delta\mu/2$, as can be seen from eqn. (4). Note that the charge current is kept at a constant (zero) value, and that the regular charge thermopower $S \Delta T$ also does not change.

A significant thermal spin accumulation is observed in the Si upon heating it (Fig. 3). The Hanle curve is identical for both directions of the Joule heating current, implying that the sign and magnitude of the spin polarization induced in the Si are the same. The peak amplitude scales quadratically with the heating current density and thus linearly with the applied heating power (right panel), as expected. The results demonstrate that a spin current is created across the tunnel contact without a net charge tunnel current. Similar experiments were done with the heating current applied to the ferromagnet, showing that the spin current (and resulting spin accumulation) change sign when the direction of the heat flow across the tunnel contact is reversed. Together with a series of control experiments to exclude artifacts,¹⁸ this established the observation of Seebeck spin tunneling.

Figure 3. Seebeck spin tunneling in $\text{Ni}_{80}\text{Fe}_{20}/\text{Al}_2\text{O}_3/\text{Si}$ tunnel contacts.¹⁸ (Top) Principle of the experiment and detailed device layout. (Bottom) Hanle data for both polarities of the Joule heating current in the Si, as well as the quadratic and linear increase of the amplitude of the spin accumulation with heating current density and power, respectively.

The experiments by Le Breton et al.¹⁸ revealed the phenomenon of Seebeck spin tunneling, and simultaneously demonstrated that it can be *used* for thermally-driven spin injection into a semiconductor. The observation of the tunnel magneto-Seebeck effect was reported very shortly thereafter in MgO-based tunnel junctions,^{26,27} following the theoretical prediction,^{7,40} which was made without connecting it to Seebeck spin tunneling (since at that time Seebeck spin tunneling was not yet known). These advances have triggered a recent surge of investigations of spin caloritronics phenomena in tunnel structures. Seebeck spin tunneling has now been reproduced for Ge devices with MgO-based tunnel contacts.^{41,42} The tunnel magneto-Seebeck effect has been studied in magnetic tunnel junctions with Al₂O₃ barriers,⁴³ and several other reports on the topic have recently appeared.^{44–47} Anisotropy of the tunnel magnetothermopower in GaMnAs based-tunnel devices was observed,⁴⁸ whereas deviations from Ohm’s law due to spin-thermoelectric coupling and Seebeck rectification up to microwave frequencies have been reported in MgO-based tunnel junctions.⁴⁹ On the theory front, ab initio studies of the tunneling magneto-Seebeck effect^{50,51} and thermally-induced spin-transfer torques in tunnel junctions⁵² have been reported, following earlier predictions of thermal torques in magnetic heterostructures.^{14,53}

5. ANISOTROPY OF THE SEEBECK COEFFICIENT

Anisotropy is a topic of significant interest since it is ultimately determined by spin-orbit interaction, which is important for many spin-related phenomena. In magnetic tunnel contacts in which only one of the electrodes is ferromagnetic, the spin-orbit interaction at the ferromagnet/insulator interface gives rise to TAMR (tunneling anisotropic magnetoresistance⁵⁴). This refers to the dependence of the tunnel resistance on the absolute orientation of the magnetization of the ferromagnet. Such an anisotropy may also appear in spin-dependent thermoelectric effects, and indeed, anisotropy of the magnetothermopower in GaMnAs based-tunnel devices was reported.⁴⁸ In magnetic tunnel contacts on silicon with an electrically created spin accumulation, anisotropy was also reported.^{55,56} Since for purely electrical spin-transport the total voltage is given by $V = R_{tun} I + P_G \Delta\mu/2$ (see eqn. (4)), the anisotropy can arise from anisotropy of the tunnel resistance (i.e., from TAMR), or from anisotropy of the induced spin accumulation. The latter can be due to anisotropy of the tunnel spin polarization P_G or the spin relaxation time in the semiconductor.

To investigate whether the Seebeck effect of a magnetic tunnel contact on silicon is anisotropic, a thermal spin accumulation was created by applying a Joule heating current to the Si, and the voltage across the tunnel contact was measured at zero net charge tunnel current. When a sufficiently large magnetic field ($B > 1\text{ T}$) is applied such that the magnetization of the ferromagnetic Ni₈₀Fe₂₀ electrode is aligned with the (in-plane or out-of-plane) field, the field and injected spins are always parallel and there is no spin precession. In the absence of anisotropy, the tunnel voltage should then be independent of the magnetization direction. Experimentally, the voltage for large in-plane magnetic field saturates at a higher level than it does for large perpendicular field (Fig. 4). We thus conclude that the Seebeck effect is different for the in-plane and out-of-plane situation.

Figure 4. Anisotropy of the Seebeck effect in a Ni₈₀Fe₂₀/Al₂O₃/Si tunnel junction with the Si heated. The anisotropy is revealed by the fact that the Hanle and inverted Hanle curves do not saturate at the same level at large applied magnetic fields for which the magnetization is aligned with the field and spin precession is absent regardless of the field orientation. Thus, the Seebeck coefficient is different for in-plane and out-of-plane magnetization. Note that both curves are taken from Ref.¹⁸ in which they were, however, not plotted together such that the anisotropy was not evident.

Compared to the previously observed anisotropy of electrical spin transport in similar devices,^{55,56} the origin of the anisotropy for thermal spin transport is different. The thermal spin current is measured at zero charge tunnel current, and the total voltage across the tunnel contact is given by $V = P_G \Delta\mu/2 + S \Delta T$ (see eqn. (4)). Since the thermally-induced $\Delta\mu$ is proportional to ΔT , the thermovoltage can be written¹⁹ in terms of a spin and a charge contribution, which together produce the observed anisotropy of the Seebeck effect. However, TAMR cannot contribute since the charge current is zero, and the term $R_{tun} I$, which is responsible for the TAMR contribution to the anisotropy, is absent for the thermal spin transport. Hence, the observed thermal anisotropy of the $\text{Ni}_{80}\text{Fe}_{20}/\text{Al}_2\text{O}_3/\text{Si}$ tunnel contacts is due to the spin and/or the charge part of the thermopower.

6. SUMMARY

The observation of Seebeck spin tunneling, the thermoelectric analog of spin-polarized tunneling, has revealed that the Seebeck coefficient of a ferromagnetic tunnel contact depends on spin. Consequently, a heat flow across a ferromagnetic tunnel contact is accompanied by a pure spin current, without a charge tunnel current. It enables the thermal transfer of spin-based information into non-magnetic materials, such as silicon, which hitherto required sending an electrical charge current into it. Seebeck spin tunneling is also the fundamental origin of the tunnel magneto-Seebeck effect in magnetic tunnel junctions. The Seebeck coefficient is found to be anisotropic, i.e., dependent on the absolute magnetization direction of the ferromagnetic electrode. Seebeck spin tunneling provides prospects for the functional use of heat and opens the way to built novel types of spin-based electronic devices and circuits for an energy efficient information technology.

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