Thermal spin current and magnetothermopower by Seebeck spin tunneling

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The recently observed Seebeck spin tunneling, the thermoelectric analog of spin-polarized tunneling, is described. The fundamental origin is the spin dependence of the Seebeck coefficient of a tunnel junction with at least one ferromagnetic electrode. Seebeck spin tunneling creates a thermal flow of spin-angular momentum across a tunnel barrier without a charge tunnel current. In ferromagnet/insulator/semiconductor tunnel junctions, this can be used to induce a spin accumulation $\Delta\mu$ in the semiconductor in response to a temperature difference ΔT between the electrodes. A phenomenological framework is presented to describe the thermal spin transport in terms of parameters that can be obtained from experiment or theory. Key ingredients are a spin-polarized thermoelectric tunnel conductance and a tunnel spin polarization with nonzero energy derivative, resulting in different Seebeck tunnel coefficients $S_{st}^{\uparrow}$ and $S_{st}^{\downarrow}$ for majority and minority spin electrons. We evaluate the thermal spin current, the induced spin accumulation and $\Delta\mu/\Delta T$, discuss limiting regimes, and compare thermal and electrical flow of spin across a tunnel barrier. A salient feature is that the thermally induced spin accumulation is maximal for smaller tunnel resistance, in contrast to the electrically induced spin accumulation that suffers from the impedance mismatch between a ferromagnetic metal and a semiconductor. The thermally induced spin accumulation produces an additional thermovoltage proportional to $\Delta\mu$, which can significantly enhance the conventional charge thermopower. Owing to the Hanle effect, the thermopower can also be manipulated with a magnetic field, producing a Hanle magnetothermopower.

I. INTRODUCTION

The interplay of heat and charge transport is the basis of thermoelectrics, enabling the conversion of heat flow to electrical power and vice versa. Spintronics concerns the interplay of spin and charge transport and has transformed magnetic data storage technology and magnetic field sensing. The connection between these two important fields has been established in studies of thermoelectric properties of magnetic nanostructures.[1–10] This interplay between heat and spin transport, now referred to as spin caloritronics,[9,11] has recently gained impetus because the combination of thermoelectrics and spintronics offers unique possibilities. 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 create thermal gradients that might influence magnetic and spin-related phenomena and thereby device performance and efficiency. This underpins the importance of understanding the fundamental interactions between thermal and spin effects.

A notable recent development is the observation of the spin Seebeck effect by Uchida *et al.*.[12] They found that when a ferromagnetic material (permalloy) is subjected to a thermal gradient ∇T, a spin current is injected into a nonmagnetic metal (Pt) strip attached to the ferromagnet. This spin current is converted into a voltage proportional to ∇T via the inverse spin Hall effect.[12] The name "spin Seebeck effect" suggests it is the spin analogue of the classical charge Seebeck effect. The latter can be understood in the following way, noting that the electrical conductance depends on the energy of the charge carriers. A thermal gradient across a (nonmagnetic)

conductor causes a flow of electrons with energy above the Fermi energy from the hot to the cold side. Simultaneously, electrons with energy below the Fermi energy flow in the opposite direction. There is a net current because the two current components do not cancel when the conductivity for electrons above and below the Fermi energy is different. In open circuit conditions, this results in a voltage between the hot and cold ends of the conductor, proportional to $S\nabla T$, with S the Seebeck coefficient. In a ferromagnetic conductor, one expects that the Seebeck coefficient is different for electrons with majority and minority spins, as their electronic properties are different. A thermal gradient across a ferromagnet would then yield a net flow of spin parallel to the thermal gradient, and produce in a spin voltage (accumulation of spin) at the hot and the cold ends of the ferromagnet. Although this was originally suggested to be the cause of the observed spin Seebeck effect,[12] the currently accepted interpretation is rather different. It is now considered to originate from a nonequilibrium between the magnon distribution in the ferromagnet and the electrons in the attached nonmagnetic metal, resulting in thermally driven dynamical spin pumping across the interface, without a global spin current or spin accumulation in the ferromagnet.[13–17] This microscopic mechanism bears no relation with the classical charge Seebeck effect. Yet, the spin Seebeck effect is a novel method to convert a thermal gradient into a voltage, via the spin, and the phenomenon is generic, i.e., subsequent to the original demonstration for permalloy,[12] it was also observed in ferromagnetic insulators,[18] ferromagnetic semiconductors,[19,20] and other ferromagnetic metals.[21]

In a different type of experiment, Slachter *et al.*[22] demonstrated for the first time that the Seebeck coefficient of a ferromagnet depends on spin. They showed that a thermal gradient in ferromagnetic permalloy induces a spin current in the permalloy parallel to the heat flow, and that when the

heat flow is directed toward an interface with a non-magnetic metal, the spin current crosses the interface and produces a spin accumulation in the nonmagnetic metal. This thermally driven spin injection is directly proportional to the difference of the Seebeck coefficient of majority- and minority-spin electrons *in* the ferromagnet, which was shown to be a fraction of the regular charge Seebeck coefficient of the ferromagnet.[22,23]

A distinctly different phenomenon, Seebeck spin tunneling, was observed by Le Breton *et al*.[24] Unlike previous work, Seebeck spin tunneling is a pure interface effect that occurs in tunnel junctions with a temperature difference ΔT between the two electrodes, provided that at least one of the electrodes is ferromagnetic. It was demonstrated that Seebeck spin tunneling creates a flow of spin angular momentum across a tunnel barrier *without* a charge tunnel current. This thermal spin current was shown to be governed by the variation of the spin polarization of the tunneling process with the energy of the tunneling electrons. As will be explained here, Seebeck spin tunneling is directly linked to the spin-dependent Seebeck coefficient of a magnetic tunnel contact. This implies that the results of Le Breton *et al*. effectively demonstrated that the Seebeck tunnel coefficient for majority and minority spin is different. In addition, Le Breton *et al*. used Seebeck spin tunneling for thermally driven spin injection into a semiconductor, i.e., they observed that in ferromagnet/insulator/silicon tunnel contacts, the thermal spin current induces a spin accumulation $\Delta \mu$ in the silicon.

An interesting analogy exists between electrical and thermal spin transport across a tunnel junction. Seebeck spin tunneling is the thermoelectric analog of spin-polarized tunneling, which refers to the spin dependence of the electrical conductance of a magnetic tunnel contact. The latter was clearly demonstrated four decades ago in experiments[25,26] on ferromagnet/insulator/superconductor junctions, showing that the charge tunnel current between a ferromagnet and a nonmagnetic counterelectrode is spin polarized. In magnetic tunnel junctions comprising of *two* ferromagnetic electrodes, spin-polarized tunneling also gives rise to large tunnel magnetoresistance,[27,28] denoting the change of the tunnel resistance as a function of the relative orientation (parallel versus antiparallel) of the magnetization of the electrodes. Analogously, Seebeck spin tunneling produces a tunnel magnetothermopower, i.e., a dependence of the thermopower of a magnetic tunnel junction on the relative magnetization alignment of the two electrodes. This tunnel magnetothermopower (or tunnel magneto-Seebeck effect) has been theoretically predicted[2,29] and recently observed by different groups, first, in MgO-based tunnel junctions[30,31] and, subsequently, in Al_2O_3 junctions.[32] Anisotropy of the tunnel magnetothermopower was also reported.[33] Last but not least, it was predicted that thermal gradients give rise to thermal spin-transfer torques in magnetic heterostructures[9,34] and tunnel junctions[35] and experimental evidence for thermal torques has been presented.[36,37]

Le Breton *et al*.[24] described the salient features of Seebeck spin tunneling by numerical evaluation of a free-electron model. Here, we present a phenomenological framework to describe Seebeck spin tunneling in linear response in terms of parameters that can be obtained from experiment and analytical or *ab initio* theory. Key ingredients are a tunnel

conductance with a spin polarization that depends on energy, and the spin polarization of the thermally induced electrical transport across the tunnel barrier. An important aim is to establish the connection with a Seebeck tunnel coefficient that depends on spin. We evaluate the thermal spin current, the induced spin accumulation and $\Delta \mu / \Delta T$, and show that these are proportional to $S_{st}^{\uparrow} - S_{st}^{\downarrow}$, where $S_{st}^{\uparrow}$ and $S_{st}^{\downarrow}$ denote the Seebeck tunnel coefficient for majority and minority spin, respectively. We discuss limiting regimes and point out that the thermally induced spin accumulation increases for smaller tunnel resistance, in contrast to the electrically induced spin accumulation that suffers from the impedance mismatch between a ferromagnetic metal and a semiconductor.[38–40] We also compare the fundamental limits of thermal and electrical spin tunneling. Finally, we demonstrate that the thermally induced spin accumulation produces an additional thermovoltage proportional to $\Delta \mu$ that can significantly enhance the conventional charge thermopower. The thermopower can be manipulated with a magnetic field owing to the Hanle effect, producing a Hanle magnetothermopower in junctions with only one ferromagnetic electrode.

II. SEEBECK SPIN TUNNELING

A. Model

We consider a tunnel junction with a ferromagnetic electrode and a nonferromagnetic electrode, typically a semiconductor (see Fig. 1) or metal. It is assumed that the tunnel resistance R_{tun} is much larger than the resistance of the electrodes, such that tunneling limits the transport. The ferromagnetic and nonmagnetic electrode are characterized by so-called spin resistances r_s^{fm} and r_s, respectively, describing the ratio of the spin accumulation in the material and the associated spin current due to spin relaxation.[40–42] The value of R_{tun} relative to r_s^{fm} and r_s plays an important role in the spin transport, as it determines the coupling between the two spin systems. We will assume that $R_{tun} \gg r_s^{fm}$, which is
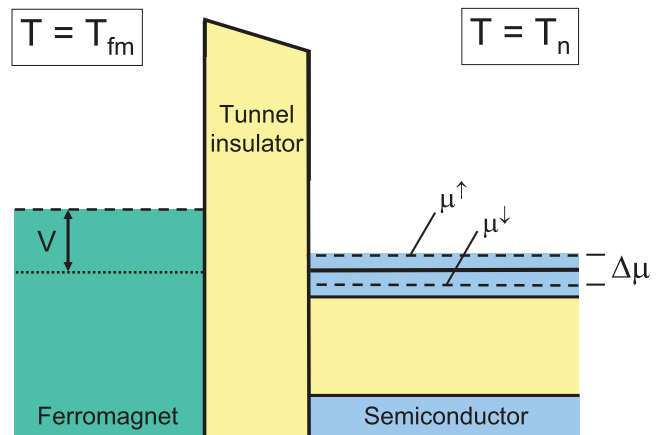

FIG. 1. (Color online) Energy band diagram of a ferromagnet/insulator/semiconductor tunnel junction. The semiconductor electrode is at temperature T_n, whereas the ferromagnet is at T_{fm}. A spin accumulation exists in the semiconductor, described by a spin splitting $\Delta \mu = \mu^{\uparrow} - \mu^{\downarrow}$ of the electrochemical potential. The applied bias voltage V is referenced to the spin-average potentials.

usually the case when an interface with a (Schottky or oxide) tunnel barrier is formed (the resistance-times-area product is 1-10 $\Omega\mu m^2$ or larger for magnetic tunnel junctions,[28,43] while $r_s^{\rm fm}$ is typically below 0.01 $\Omega\mu m^2$ for transition-metal ferromagnets[40–42]). Therefore, we do not need to consider the spin accumulation and spin-dependent (electrical or thermal) transport *within* the ferromagnet, including the spin-dependent Seebeck effect[22,23] due to any temperature gradients within the ferromagnet (this is important for the regime $R_{\rm tun} < r_s^{\rm fm}$, as recently shown[44]). The spin resistance of the nonmagnetic material is normally much larger than $r_s^{\rm fm}$. We do not make any specific assumptions about the value of $R_{\rm tun}$ relative to r_s. We thus cover the regime with $R_{\rm tun} > r_s$ where the spin accumulation in the semiconductor is effectively decoupled from the ferromagnet by the tunnel barrier as well as the regime with $R_{\rm tun} < r_s$ where the coupling to the ferromagnet reduces the spin accumulation in the nonmagnetic material.[38–40]

For a given bias voltage V and temperature difference ΔT across the barrier, the tunnel current I^σ for each spin ($\sigma = \uparrow, \downarrow$, denoting electrons with magnetic moment, respectively, parallel and antiparallel to the magnetization of the ferromagnet) is

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

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

The first term on the right-hand side describes the electrically driven current governed by spin-dependent tunnel conductances $G^\uparrow$ and $G^\downarrow$. It incorporates the effect of the shifts of the electrochemical potentials μ^σ in the semiconductor due to the presence of the spin accumulation $\Delta\mu = \mu^\uparrow - \mu^\downarrow$ (for convenience we have defined $\Delta\mu$ in units of volt). Note that the spin accumulation typically decays exponentially with distance from the injection contact and that $\Delta\mu$ denotes the value of the spin accumulation at the interface with the tunnel barrier, as is relevant for tunneling. The second term on the right-hand side describes the thermally induced tunnel current in response to a temperature difference, as governed by $L^\uparrow$ and $L^\downarrow$, which we will refer to as the thermoelectric tunnel conductances (not to be confused with the thermal conductance that describes heat flow). We define $\Delta T = T_n - T_{\rm fm}$, where T_n and $T_{\rm fm}$ are the temperatures of the nonmagnetic and ferromagnetic electrode, respectively, and $V = V_n - V_{\rm fm}$, where V_n and $V_{\rm fm}$ are the spin-averaged potentials of the nonmagnetic and ferromagnetic electrode.

The total conductances are $G = G^\uparrow + G^\downarrow$ and $L = L^\uparrow + L^\downarrow$, and their spin polarizations are $P_G = (G^\uparrow - G^\downarrow)/(G^\uparrow + G^\downarrow)$ and $P_L = (L^\uparrow - L^\downarrow)/(L^\uparrow + L^\downarrow)$. The charge tunnel current I and the spin tunnel current I_s are then

$$I = I^\uparrow + I^\downarrow = G V - P_G G \left(\frac{\Delta\mu}{2} \right) + L \Delta T, \qquad (3)$$

$$I_s = I^\uparrow - I^\downarrow = P_G G V - G \left(\frac{\Delta\mu}{2} \right) + P_L L \Delta T. \qquad (4)$$

The spin current consists of an electrical ($P_G G V$) and a thermal contribution ($P_L L \Delta T$) as well as a correction due to the $\Delta\mu$ that is induced by the (electrical and/or thermal) spin current. The feedback of $\Delta\mu$ on the spin/charge tunnel current implies that another (independent) relation between $\Delta\mu$ and I_s is required to obtain a solution. This is provided by the requirement of a steady-state spin accumulation in the nonmagnetic material, which implies that the spin current I_s injected by tunneling is balanced by the spin current due to spin relaxation in the material, integrated over its full spatial extent. The spin current associated with spin relaxation is proportional to the spin accumulation. We define a spin resistance r_s of the nonmagnetic material via

$$\Delta\mu = 2 I_s r_s. \qquad (5)$$

In our model, r_s is a phenomenological parameter that describes the conversion of the spin current I_s that is injected by tunneling, into a spin accumulation $\Delta\mu$. As mentioned before, $\Delta\mu$ denotes the value of the spin accumulation right at the tunnel interface. This definition of r_s makes no specific assumptions about the spatial profile of the spin accumulation in the nonmagnetic material, or the formalism used to compute it. If we use the spin-diffusion equation and a spin accumulation that decays exponentially with distance from the injection interface with the spin-diffusion length $L_{\rm sd}$, then the spin resistance of a unit contact area can be expressed as $\rho_n L_{\rm sd}$, where ρ_n is the resistivity of the nonmagnetic material.[40–42] This result is frequently used to analyze experimental data, but note that it requires introduction of a somewhat unusual factor of two in Eq. (5). If $r_s = \rho_n L_{\rm sd}$ is used, our result is consistent with a recent evaluation based on the spin-diffusion equation.[44]

B. Spin current and Seebeck spin tunnel coefficient

Equations (3)–(5) fully define the system and allow us to obtain the relevant quantities. We first derive a general expression for the spin accumulation, valid for electrical ($I \neq 0$) and thermal ($\Delta T \neq 0$) injection as well as for a combination of the two. We shall discuss the case of purely thermal ($I = 0$) and purely electrical ($\Delta T = 0$) driving force later on. The solutions for $\Delta\mu$ and the spin current in terms of ΔT and I are

$$\Delta\mu = \left[\frac{2 r_s}{R_{\rm tun} + (1 - P_G^2) r_s} \right] \times [(P_G) R_{\rm tun} I - (P_L - P_G) S_0 \Delta T], \qquad (6)$$

$$I_s = \left[\frac{1}{R_{\rm tun} + (1 - P_G^2) r_s} \right] \times [(P_G) R_{\rm tun} I - (P_L - P_G) S_0 \Delta T], \qquad (7)$$

where $R_{\rm tun} = 1/G$ is the tunnel resistance and $S_0 = -L/G$ is the charge thermopower (in the absence of a spin accumulation; see below). The first term in Eq. (7), proportional to I, is the electrical spin current associated with the spin-polarized charge current. The second term is the pure spin current due to Seebeck spin tunneling (driven by ΔT) and will be referred to as the Seebeck spin current. The thermal spin current and the corresponding spin accumulation are odd functions of ΔT and thus change sign when the sign of the temperature difference is reversed, as observed in experiment.[24] The Seebeck spin tunneling coefficient $S_{\rm st} = \Delta\mu/\Delta T$ is obtained by setting $I = 0$ in Eq. (6), and rewriting $(P_L - P_G) S_0$ in terms of spin-dependent Seebeck tunnel coefficients $S_{\rm st}^\uparrow = -L^\uparrow/G^\uparrow$ and

$S_{\text{st}}^{\downarrow} = -L^{\downarrow}/G^{\downarrow}$ for majority and minority spin, respectively. We then obtain an important result, namely, $\Delta\mu/\Delta T$ is proportional to $(S_{\text{st}}^{\uparrow} - S_{\text{st}}^{\downarrow})$:

$$S_{\text{st}} = \frac{\Delta\mu}{\Delta T} = \left[\frac{\left(1 - P_G^2\right) r_s}{R_{\text{tun}} + \left(1 - P_G^2\right) r_s}\right](S_{\text{st}}^{\uparrow} - S_{\text{st}}^{\downarrow}). \quad (8)$$

Since $0 \leqslant P_G^2 \leqslant 1$, the prefactor always has a positive sign. The sign of S_{st} is thus determined by the difference between $S_{\text{st}}^{\uparrow}$ and $S_{\text{st}}^{\downarrow}$. Note that the prefactor tends to zero when the tunnel spin polarization becomes very large ($P_G \approx 1$ or -1), but S_{st} does not because also G^{σ} for one of the two spin channels goes to zero. Hence either $S_{\text{st}}^{\uparrow}$ or $S_{\text{st}}^{\downarrow}$ diverges. Taking this into account, one finds that $S_{\text{st}} = +4\,(L^{\downarrow}/G^{\uparrow})(r_s/R_{\text{tun}})$ for $P_G = 1$ and $S_{\text{st}} = -4\,(L^{\uparrow}/G^{\downarrow})(r_s/R_{\text{tun}})$ for $P_G = -1$. Expressions (6) and (7) apply to situations for which I and ΔT are fixed. This includes recent experiments on Seebeck spin tunneling[24] (where $I = 0$) as well as most experiments on electrical spin injection that are performed in constant current mode (and $\Delta T = 0$). Alternatively, we can express $\Delta\mu$ and I_s in terms of ΔT and V. However, care has to be taken not to set $V = I\,R_{\text{tun}}$, as this is not correct when $\Delta\mu \neq 0$ or $\Delta T \neq 0$, see Eq. (3).

C. Charge thermopower and Hanle magnetothermopower

The charge thermopower S is obtained from the voltage $V|_{I=0}$ for which I vanishes. From Eq. (3), we obtain

$$S = \frac{V|_{I=0}}{\Delta T} = S_0 + \left(\frac{P_G}{2}\right) S_{\text{st}}. \quad (9)$$

This is another important result. In the presence of a spin accumulation, the charge thermopower is not equal to $S_0 = -L/G$. There is an additional, previously unidentified, contribution that is proportional to $\Delta\mu$ and thus to the Seebeck spin tunnel coefficient S_{st}. Since P_G can be positive or negative depending on the properties of the ferromagnet/insulator interface, and also S_{st} can have either sign, the additional contribution can enhance or reduce the charge thermopower. The enhancement can be significant because S_{st} can be much larger than S_0, as we will see in the discussion section.

Next, we address how the thermally induced spin accumulation can be detected as a voltage signal. Just as for electrically induced spin accumulation, this can be done via the Hanle effect, which occurs when the spins in the semiconductor are subjected to a magnetic field B at a solid angle θ with the spin direction.[45–47] This causes spin precession and consequently a reduction of $\Delta\mu$ depending on θ and on the product of the spin lifetime τ_s and the Larmor frequency $\omega_L = g\mu_B B/\hbar$, where g is the Landé g factor, μ_B the Bohr magneton, and $\hbar$ is the Planck's constant divided by 2π. When the tunnel resistance is sufficiently large ($R_{\text{tun}} \gg r_s$) such that the coupling of the spin accumulation to the ferromagnet can be neglected, spin precession causes a decay of $\Delta\mu$ in a Lorentzian fashion:[45,47]

$$\Delta\mu = 2\,I_s\,r_s \equiv 2\,I_s\,r_s^0 \left[\cos^2(\theta) + \frac{\sin^2(\theta)}{1 + (\omega_L\tau_s)^2}\right]. \quad (10)$$

In the absence of any magnetic field, there is no spin precession and the spin resistance is r_s^0. If we keep ΔT and the charge current I constant, apply a magnetic field perpendicular to the spins, and increase B from zero to a value for which $\omega_L\tau_s \gg 1$,

the spin resistance is gradually reduced from r_s^0 to zero. The $\Delta\mu$ then also goes to zero, even if the spin current I_s that is injected by tunneling is nonzero. This results in the desired voltage change, which is obtained from Eqs. (3) and (10) as

$$\Delta V_{\text{Hanle}} = V|_{\omega_L=0} - V|_{\omega_L\tau_s\gg1} = \left(\frac{P_G}{2}\right)\Delta\mu|_{\omega_L=0}. \quad (11)$$

An important point is that this expression is valid irrespective of how the spin accumulation is created. In other words, also for a thermally induced spin accumulation, the detected voltage signal ΔV_{Hanle} is given by $P_G/2$ times $\Delta\mu$, which is the same relation as for electrically induced spin accumulation.[46]

In the regime where $R_{\text{tun}} < r_s$, the magnitude of $\Delta\mu$ is reduced by the coupling of the spins to the ferromagnet, but also the functional dependence of $\Delta\mu$ on B is modified and Eq. (10) does not correctly describe the dependence on B and θ. However, the maximum and minimum values of $\Delta\mu$ for, respectively, $\omega_L = 0$ and $\omega_L\tau_s \gg 1$ are still properly described. Therefore the amplitude of the spin accumulation can still be correctly obtained from Eq. (11). However, extracting the spin lifetime from the linewidth of the Hanle curve requires a detailed description of the line shape taking the interaction with the ferromagnet into account.

The ability to manipulate the spin accumulation with an external magnetic field (owing to the Hanle effect) also means that S_{st} and hence the charge thermopower S can be controlled by a magnetic field. We define the Hanle magnetothermopower S_{mag} as the relative change of the thermopower between its value in zero magnetic field, and the value at $\Delta\mu = 0$ that corresponds to $\omega_L\tau_s \gg 1$:

$$S_{\text{mag}} = \frac{S|_{\omega_L=0} - S|_{\omega_L\tau_s\gg1}}{S|_{\omega_L\tau_s\gg1}} = \left(\frac{P_G}{2}\right)\left(\frac{S_{\text{st}}|_{\omega_L=0}}{S_0}\right). \quad (12)$$

The magnetothermopower is mediated by the spin accumulation, has a variation with magnetic field that is governed by the Hanle effect, and occurs in tunnel contacts in which only one of the electrodes is ferromagnetic. It is thus different from the tunnel magnetothermopower recently observed in a magnetic tunnel junction with two ferromagnetic electrodes, which has a magnetic field variation that is controlled by the angle between the magnetizations of the two electrodes.[30,31] The Hanle magnetothermopower can be very large because S_{st} can be much larger than S_0. The field scale for the Hanle magnetothermopower is set by the spin lifetime. The half-width (for which $\omega_L\tau_s = 1$) is inversely proportional to τ_s, and for a typical value of $\tau_s = 1$ ns, the half width is about 60 Gauss (6 mT) for a nonmagnetic material with a g factor of 2.

III. DISCUSSION

A. Origin and definition of Seebeck spin tunneling

We have seen that Seebeck spin tunneling occurs when the Seebeck coefficient of a magnetic tunnel contact is different for majority and minority spin. Le Breton *et al.*[24] described the salient features of Seebeck spin tunneling by numerical evaluation of a free electron model, and showed that Seebeck spin tunneling is determined by the energy derivative of the tunnel spin polarization. In this section, we will establish the

important connection between these two notions, and also clarify the definition of Seebeck spin tunneling.

First, we note that $(S_{st}^{\uparrow} - S_{st}^{\downarrow})$ is proportional to $(P_L - P_G)$ and that the spin accumulation induced by Seebeck spin tunneling is proportional to $(P_L - P_G)$. This is easily understood because when $I = 0$, any thermally induced current (with polarization P_L) must be balanced by an equal but opposite electrically driven current (with polarization P_G). If the tunnel spin polarization does *not depend on energy*, all the induced (electrical or thermal) current components necessarily have the same spin polarization, and we have $P_L = P_G$. In that case, $S_{st} = 0$, the Seebeck spin current vanishes for all ΔT, and a spin current exists only if the charge current is nonzero. To illustrate that $P_L = P_G$ if the tunnel spin polarization does not depend on energy, we express G^{σ} and L^{σ} in terms of the tunnel transmission function $D^{\sigma}(E)$ integrated over energy E, as[29,48]

$$G^{\sigma} = -\frac{e^2}{h} \int D^{\sigma}(E) \left[\partial_E f(E, \mu, T) \right] dE, \qquad (13)$$

$$L^{\sigma} = -\frac{e}{h}\frac{1}{T} \int D^{\sigma}(E)(E - \mu) \left[\partial_E f(E, \mu, T) \right] dE, \quad (14)$$

where $\partial_E f(E, \mu, T)$ is the energy derivative of the Fermi-Dirac distribution function $f(E, \mu, T)$. When $D^{\uparrow}(E)$ and $D^{\downarrow}(E)$ have the same variation with energy, we can write $D^{\sigma}(E) = \chi^{\sigma} D(E)$, where the coefficients $\chi^{\uparrow}$ and $\chi^{\downarrow}$ do not depend on energy. Inserting this in Eqs. (13) and (14), we find $P_G = P_L = (\chi^{\uparrow} - \chi^{\downarrow})/(\chi^{\uparrow} + \chi^{\downarrow})$. Then P_G and P_L are independent of E and $P_L = P_G$. Since $(S_{st}^{\uparrow} - S_{st}^{\downarrow})$ is proportional to $(P_L - P_G)$, we conclude that $S_{st}^{\uparrow} = S_{st}^{\downarrow}$ if the tunnel spin polarization does not depend on energy. This establishes the connection between the energy derivative of P_G and a spin-dependent Seebeck coefficient: $S_{st}^{\uparrow} \neq S_{st}^{\downarrow}$ only if the tunnel spin polarization depends on energy.

We now define the term Seebeck spin tunneling more precisely, because one could argue that a thermally driven spin current can exist even if $P_L = P_G$ or, equivalently, if $S_{st}^{\uparrow} = S_{st}^{\downarrow}$. This can be seen more easily by writing Eq. (7) in terms of applied bias V:

$$I_s = \left[\frac{1}{R_{\text{tun}} + r_s} \right] \left[P_G \left(V - S_0 \Delta T \right) - (P_L - P_G) S_0 \Delta T \right]. \tag{15}$$

When $P_L = P_G$, a term proportional to $P_G S_0 \Delta T$ is still present. This term, which was already discussed by Johnson and Silsbee,[1] originates from the thermally induced charge current, which is spin polarized due to the nonzero value of P_G. In fact, for $P_L = P_G$ any thermally induced spin current is from the spin-polarized charge current that arises from the shift of the I-V curve by an amount equal to the charge thermovoltage $S_0 \Delta T$. Although technically this is a thermally-induced spin current, the charge current is nonzero, i.e., it is not a pure spin current, as can be seen from Eq. (7). Since this spin current exists even if the tunnel spin polarization is not dependent on energy, we do not consider this to be Seebeck spin tunneling. Seebeck spin tunneling thus refers to the other term, proportional to $(P_L - P_G)$, that is associated with a nonzero energy derivative of the tunnel spin polarization (in analogy with the conventional charge Seebeck effect that is related

to a nonzero energy derivative of the charge conductivity). The Seebeck spin tunneling term and its relation with the energy dependence of P_G has not been discussed or described in previous theoretical treatments of thermally driven spin transport across interfaces.[1,10] Experimentally, one will have a combination of a thermally driven spin-polarized charge current and Seebeck spin tunneling, unless one measures at $I = 0$, as done in Ref. 24.

There is ample evidence for the energy dependence of the tunnel spin polarization. Indirect evidence comes from the decay of tunnel magnetoresistance with bias voltage in magnetic tunnel junctions,[27,28] which is, in part, governed by the decay of the tunnel spin polarization with energy. Direct evidence is provided in two reports for transition metal ferromagnets on Al_2O_3, where the variation of the tunnel spin polarization with energy of the tunnel electrons was determined.[49,50] A significant asymmetry in the decay of the tunnel spin polarization with energy below and above the Fermi energy was reported, the decay being much faster above the Fermi energy.

B. Comparison of electrical and thermal spin current

It is instructive to compare the magnitude of the spin current due to electrical and thermal spin tunneling. Besides the fundamental interest, this is of course important from a technological point of view. One question is whether the creation of a spin current by a temperature difference across the tunnel barrier can be more energy efficient than creating a spin current electrically. Another question is whether the heat that is produced by electrical generation of a spin current can be reused to supplement it with a thermal spin current, and how much increase in spin current, or reduction in energy consumption, can be obtained in this way. The answer to those questions cannot be given in general terms. The efficiency of creating the temperature difference depends crucially on the thermal design of the structures. Moreover, whereas it has been known for four decades that the electrical tunnel conductance is spin polarized and the polarization has been rather well optimized, the Seebeck spin tunneling has only recently been observed and the difference $(S_{st}^{\uparrow} - S_{st}^{\downarrow})$ is far from optimum. We will therefore only discuss the factors that determine the ultimate limits of electrical and thermal spin current.

We consider the driving term as well as the proportionality factor between $\Delta \mu$ and the driving term (see Table I). The thermal driving term is $S_0 \Delta T$, which should be compared to the electrical driving term $R_{\text{tun}} I$. For nonmagnetic metal tunnel junctions S_0 has been evaluated[51,52] to be in the range of 50–100 μV/K, although it has been predicted that it can be enhanced by magnons in ferromagnetic tunnel junctions.[3,4] The ΔT for tunnel junctions will, in practice, be limited to about 10 K. Hence $S_0 \Delta T$ is of the order of 1 mV, which is to be compared to typical values of a few 100 mV for $R_{\text{tun}} I$. In general, the thermal driving term will thus be smaller than the electrical driving term.

With respect to the proportionality factor, for electrical spin injection it is limited by P_G, since its absolute value cannot be larger than one (by definition). However, such a restriction does not exist for the proportionality factor of the thermal spin current, since there is no limit for the energy derivatives that

TABLE I. Comparison of thermal and electrical spin current in a tunnel junction.

Method	Type of spin current	Driving term	Typical values	Proportionality	Extreme values
Electrical	Spin-polarized charge current	$R_{tun} I$	~ 100 mV	P_G	± 1
Thermal	Pure spin current ($I = 0$)	$S_0 \Delta T$	~ 0.1–1 mV	$S_{st}^{\uparrow} - S_{st}^{\downarrow}$	$\pm \infty$

govern ($S_{st}^{\uparrow} - S_{st}^{\downarrow}$) and ($P_L - P_G$). In principle, $L^{\uparrow}$ and $L^{\downarrow}$ can be equal in magnitude but of opposite sign, so that $L \approx 0$ and P_L goes to infinity. Physically, this corresponds to the situation where the tunnel spin polarizations for states above and below the Fermi energy have opposite sign, such that one type of spin is driven from the hot to the cold side of the tunnel contact, and the other type of spin is driven from the cold to the hot side. Hence the proportionality factor for thermal spin accumulation can, in principle, be arbitrarily large for suitably engineered materials. This can therefore (more than) compensate for the smaller thermal driving term. This suggests that Seebeck spin tunneling can be a viable approach to create a spin current, either by itself, or in conjunction with an electrical spin current.

C. Magnitude of Seebeck spin tunnel coefficient

The magnitude of the thermal spin current (and spin accumulation) depends on the value of the polarizations P_L and P_G as well as on the coupling of the spin accumulation to the ferromagnet. An important point is that the Seebeck spin tunnel coefficient S_{st} can be much larger than the regular charge thermopower S_0. To illustrate this, the ratio of S_{st} and S_0 is shown as a function of relevant parameters in Fig. 2. For materials with large tunnel spin polarization ($P_G \approx 1$ or -1), a very large Seebeck spin tunneling coefficient is produced if P_L and P_G are unequal, or preferably, of opposite sign. This situation would occur for ferromagnet/insulator interfaces that have an almost full spin polarization of the
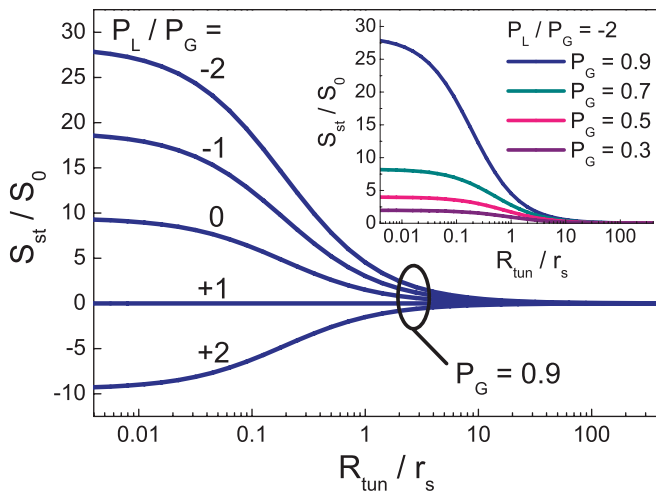
tunnel conductance for states at and below the Fermi energy E_F, but a rapidly decaying or even opposite spin polarization above E_F, for instance, due to the onset of a contribution to the tunneling of a band with opposite spin orientation. Since the total thermopower is given by the sum of S_0 and $(P_G/2) S_{st}$ [see Eq. (9)], the thermally induced spin accumulation in the nonmagnetic material can significantly enhance the charge thermopower of a tunnel junction.

D. Scaling with tunnel resistance

A noteworthy difference between electrical and thermal creation of a spin accumulation is the scaling with tunnel resistance (see Fig. 3 and Appendix A for explicit expressions for the different regimes). For electrical spin injection, the polarization of the injected current (I_s/I) is P_G as long as the tunnel resistance is larger than the spin resistance of the semiconductor [see Eq. (A4)]. However, when $R_{tun} \ll$

FIG. 2. (Color online) Seebeck spin tunnel coefficient S_{st} normalized to S_0, as a function of R_{tun}/r_s, for various values of P_L/P_G and fixed $P_G = 0.9$, and (inset) for fixed $P_L/P_G = -2$ and P_G varied. Note that $S_{st} = 0$ if $P_L/P_G = +1$.
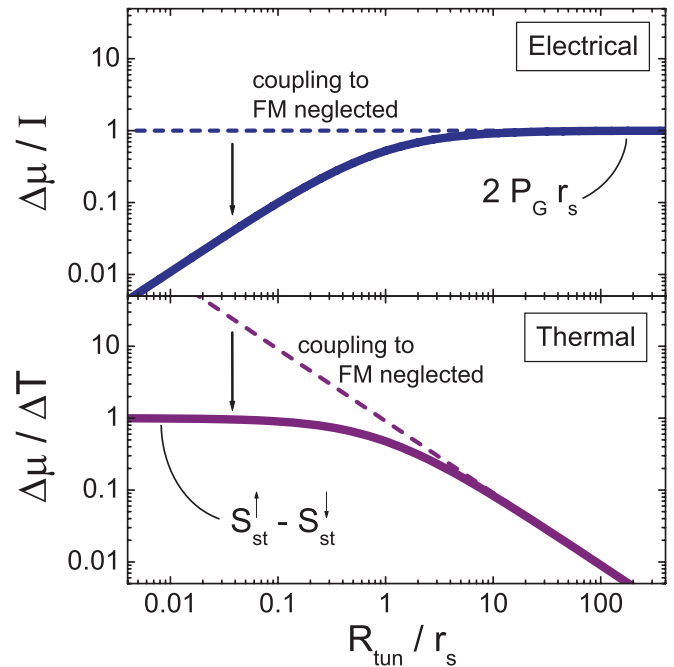

FIG. 3. (Color online) Scaling of the electrical and thermal spin accumulations with tunnel resistance. Shown are $\Delta \mu / I$ for electrical injection (top panel) and $\Delta \mu / \Delta T$ for Seebeck spin tunneling (bottom panel), both as a function of the ratio R_{tun}/r_s of the tunnel resistance and the spin resistance of the semiconductor. The results are normalized to the maximum value as indicated ($2 P_G r_s$ for electrical and $S_{st}^{\uparrow} - S_{st}^{\downarrow}$ for thermal). The dashed lines describe the result when one neglects the feedback of $\Delta \mu$ on the spin current injected from the ferromagnet. The arrows indicate the reduction due to the feedback. The P_G was set to 0.3.

r_s, the coupling to the ferromagnet starts to play a role, and the feedback of $\Delta\mu$ on the tunnel transport severely reduces the spin polarization of the tunnel current (which is well established[38–40]). As a result, $\Delta\mu/I$, the spin accumulation per unit injected charge current, is constant at large R_{tun} but decays at small tunnel resistance (see Fig. 3, top panel).

In contrast, the scaling of the thermally induced spin accumulation is opposite; $\Delta\mu/\Delta T$ *increases* as the tunnel resistance is lowered, and reaches a large and constant value when R_{tun} becomes *smaller* than r_s (see Fig. 3, bottom panel). This behavior is consistent with that obtained by numerical evaluation of a free-electron model.[24] It thus appears that thermal injection is more efficient at small tunnel resistance, whereas electrical creation of spin accumulation requires sufficiently large tunnel resistance to overcome the impedance mismatch. Note that in Fig. 3, we have neglected that S_0 decays for thinner tunnel barriers, because the decay is known to be very weak[51,52] and does not critically affect the main scaling trend. It is also known that for an ultrathin tunnel contact, the thermal (heat) conductance $I_Q/\Delta T$, with I_Q the heat current, is limited by the interfaces rather than the bulk thermal heat conductance of the tunnel barrier material. Therefore the thermal conductance $I_Q/\Delta T$ is expected to be approximately independent of R_{tun}. A similar trend would thus result if we would plot $\Delta\mu/I_Q$ instead of $\Delta\mu/\Delta T$. We thus find that the spin accumulation per unit *charge* current is maximum for *large* tunnel resistance, whereas the spin accumulation per unit *heat* current across the tunnel barrier is maximum at *small* tunnel resistance.

IV. SUMMARY

A phenomenological framework has been presented to describe Seebeck spin tunneling, the thermoelectric analog of spin-polarized tunneling. It was established that Seebeck spin tunneling originates from the spin dependence of the Seebeck coefficient of a tunnel junction with a ferromagnetic electrode, i.e., $S_{st}^{\uparrow} \neq S_{st}^{\downarrow}$. The connection with a tunnel spin polarization P_G that depends on energy was also made. Seebeck spin tunneling creates a thermal flow of spin-angular momentum across a tunnel barrier without a charge tunnel current. In ferromagnet/insulator/semiconductor tunnel junctions, it allows creation of a spin accumulation $\Delta\mu$ in the semiconductor by a temperature difference ΔT between the electrodes. We expressed the thermal spin current, the induced spin accumulation and $\Delta\mu/\Delta T$ in terms of the spin-dependent Seebeck coefficients, tunnel resistance and spin resistance of the nonmagnetic electrode. The thermally induced spin accumulation produces an additional thermovoltage proportional to $\Delta\mu$, which can significantly enhance the conventional charge thermopower. Because the spin accumulation can be manipulated via the Hanle effect, the thermopower depends on a magnetic field, producing a Hanle magnetothermopower in junctions in which only one of the electrodes is a ferromagnet. The thermally induced spin accumulation was shown to be maximum for smaller tunnel resistance, in contrast to the electrically induced spin accumulation that suffers from the impedance mismatch between a ferromagnetic metal and a semiconductor. While the efficiency of electrical spin injection

is limited by the fact that $|P_G| \leqslant 1$, no such restriction exists for thermal spin current that is determined by the energy derivative of P_G, which is unbounded. With suitably engineered materials, Seebeck spin tunneling is thus a viable option for efficient creation of spin current.

APPENDIX: SPIN CURRENT AND ACCUMULATION IN LIMITING REGIMES

1. Thermal spin current

There are two limiting regimes for Seebeck spin tunneling ($I = 0$). When $R_{tun} \gg r_s$, the induced $\Delta\mu$ remains relatively small and the feedback of $\Delta\mu$ on the tunnel transport is negligible. For this regime, one obtains

$$\frac{\Delta\mu}{\Delta T} \approx -2\left(P_L - P_G\right)\left(\frac{r_s}{R_{tun}}\right) S_0$$

$$= \left(1 - P_G^2\right)\left(\frac{r_s}{R_{tun}}\right)(S_{st}^{\uparrow} - S_{st}^{\downarrow}). \quad \text{(A1)}$$

The spin accumulation decays at larger tunnel resistance since for a tunnel contact $S_0 = -L/G$ depends only weakly on R_{tun}. This is because all (thermal or electrical) tunnel current components, and hence L and G, decay exponentially with tunnel barrier width and height.[51,52] When $R_{tun}/r_s < (1 - P_G^2)$, we have

$$\frac{\Delta\mu}{\Delta T} \approx \frac{-2\left(P_L - P_G\right)}{1 - P_G^2} S_0 = (S_{st}^{\uparrow} - S_{st}^{\downarrow}). \quad \text{(A2)}$$

In this regime, which corresponds to tunnel contacts with sufficiently low-resistance area product, S_{st} and $\Delta\mu$ do not directly depend on R_{tun}.

2. Electrical spin current

For comparison, the spin accumulation $\Delta\mu^{el}$ induced by electrical injection of a spin-polarized charge current, without a temperature difference across the tunnel barrier, is given by

$$\Delta\mu^{el} = \frac{2\,r_s\,R_{tun}}{R_{tun} + \left(1 - P_G^2\right) r_s} P_G I, \quad \text{(A3)}$$

and the spin current is

$$I_s^{el} = \frac{R_{tun}}{R_{tun} + \left(1 - P_G^2\right) r_s} P_G I. \quad \text{(A4)}$$

We remark that it is customary[39–42] to replace the real tunnel resistance R_{tun} by $(1 - P_G^2) r_B^*$, introducing r_B^* as an effective tunnel resistance. The prefactor then takes a more simple form without the factor $(1 - P_G^2)$. As a result, it is no longer evident that in order to determine the transition into the regime where the feedback of $\Delta\mu$ on the tunneling becomes relevant, one has to compare the tunnel resistance to $(1 - P_G^2) r_s$. The transition thus depends on the value of P_G. We therefore choose to retain the term $(1 - P_G^2)$ explicitly.

[1]M. Johnson and R. H. Silsbee, Phys. Rev. B **35**, 4959 (1987).

[2]Z.-C. Wang, G. Su, and S. Gao, Phys. Rev. B **63**, 224419 (2001).

[3]E. McCann and V. I. Fal'ko, Phys. Rev. B **66**, 134424 (2002).

[4]E. McCann and V. I. Fal'ko, J. Magn. Magn. Mater. **268**, 123 (2004).

[5]A. Fukushima, K. Yamagi, A. A. Tulapurkar, Y. Suzuki, H. Kubota, A. Yamamoto, and S. Yuasa, Jpn. J. Appl. Phys. **44**, L12 (2005).

[6]A. Fukushima, H. Kubota, A. Yamamoto, Y. Suzuki, and S. Yuasa, IEEE Trans. Magn. **41**, 2571 (2005).

[7]L. Gravier, S. Serrano-Guisan, F. Reuse, and J. Ph. Ansermet, Phys. Rev. B **73**, 024419 (2006).

[8]L. Gravier, S. Serrano-Guisan, F. Reuse, and J.Ph. Ansermet, Phys. Rev. B **73**, 052410 (2006).

[9]M. Hatami, G. E. W. Bauer, Q. Zhang, and P. J. Kelly, Phys. Rev. Lett. **99**, 066603 (2007).

[10]M. Hatami, G. E. W. Bauer, Q. Zhang, and P. J. Kelly, Phys. Rev. B **79**, 174426 (2009).

[11]G. E. W. Bauer, A. H. MacDonald, and S. Maekawa, Solid State Commun. **150**, 459 (2010).

[12]K. Uchida *et al*., Nature (London) **455**, 778 (2008).

[13]J. Xiao, G. E. W. Bauer, K. C. Uchida, E. Saitoh, and S. Maekawa, Phys. Rev. B **81**, 214418 (2010).

[14]J. Sinova, Nat. Mater. **9**, 880 (2010).

[15]H. Adachi, K. Uchida, E. Saitoh, J. Ohe, S. Takahashi, and S. Maekawa, Appl. Phys. Lett. **97**, 252506 (2010).

[16]H. Adachi, J. I. Ohe, S. Takahashi, and S. Maekawa, Phys. Rev. B **83**, 094410 (2011).

[17]K. Uchida, T. Ota, T. Nonaka, Y. Kajiwara, H. Adachi, S. Maekawa, J. Xiao, G. E. W. Bauer, and E. Saitoh, e-print arXiv:1111.3036v1.

[18]K. Uchida *et al*., Nat. Mater. **9**, 894 (2010).

[19]C. M. Jaworski, J. Yang, S. Mack, D. D. Awschalom, J. P. Heremans, and R. C. Myers, Nat. Mater. **9**, 898 (2010).

[20]C. M. Jaworski, J. Yang, S. Mack, D. D. Awschalom, R. C. Myers, and J. P. Heremans, Phys. Rev. Lett. **106**, 186601 (2011).

[21]S. Bosu, Y. Sakuraba, K. Uchida, K. Saito, T. Ota, E. Saitoh, and K. Takanashi, Phys. Rev. B **83**, 224401 (2011).

[22]A. Slachter, F. L. Bakker, J. P. Adam, and B. J. van Wees, Nat. Phys. **6**, 879 (2010).

[23]A. Slachter, F. L. Bakker, and B. J. van Wees, Phys. Rev. B **84**, 174408 (2011).

[24]J. C. Le Breton, S. Sharma, H. Saito, S. Yuasa, and R. Jansen, Nature (London) **471**, 82 (2011).

[25]P. M. Tedrow and R. Meservey, Phys. Rev. Lett. **26**, 192 (1971).

[26]R. Meservey and P. M. Tedrow, Phys. Rep. **238**, 173 (1994).

[27]J. S. Moodera, L. R. Kinder, T. M. Wong, and R. Meservey, Phys. Rev. Lett. **74**, 3273 (1995).

[28]S. Yuasa and D. D. Djayaprawira, J. Phys. D: Appl. Phys. **40**, R337 (2007).

[29]M. Czerner, M. Bachmann, and C. Heiliger, Phys. Rev. B **83**, 132405 (2011).

[30]M. Walter *et al.*, Nat. Mater. **10**, 742 (2011).

[31]N. Liebing, S. Serrano-Guisan, K. Rott, G. Reiss, J. Langer, B. Ocker, and H. W. Schumacher, Phys. Rev. Lett. **107**, 177201 (2011).

[32]W. Lin, M. Hehn, L. Chaput, B. Negulescu., S. Andrieu, F. Montaigne, and S. Mangin, e-print arXiv:1109.3421v2.

[33]Ts. Naydenova, P. Dürrenfeld, K. Tavakoli, N. Pégard, L. Ebel, K. Pappert, K. Brunner, C. Gould, and L. W. Molenkamp, Phys. Rev. Lett. **107**, 197201 (2011).

[34]J. C. Slonczewski, Phys. Rev. B **82**, 054403 (2010).

[35]X. Jia, K. Xia, and G. E. W. Bauer, Phys. Rev. Lett. **107**, 176603 (2011).

[36]H. Yu, S. Granville, D. P. Yu, and J. Ph. Ansermet, Phys. Rev. Lett. **104**, 146601 (2010).

[37]E. Padrón-Hernández, A. Azevedo, and S. M. Rezende, Phys. Rev. Lett. **107**, 197203 (2011).

[38]G. Schmidt, D. Ferrand, L. W. Molenkamp, A. T. Filip, and B. J. van Wees, Phys. Rev. B **62**, R4790 (2000).

[39]E. I. Rashba, Phys. Rev. B **62**, R16267 (2000).

[40]A. Fert and H. Jaffrès, Phys. Rev. B **64**, 184420 (2001).

[41]A. Fert, J.-M. George, H. Jaffrès, and R. Mattana, IEEE Trans. Elec. Dev. **54**, 921 (2007).

[42]H. Jaffrès, J.-M. George, and A. Fert, Phys. Rev. B **82**, 140408 (2010).

[43]K. Yakushiji, T. Saruya, H. Kubota, A. Fukushima, T. Nagahama, S. Yuasa, and K. Ando, Appl. Phys. Lett. **97**, 232508 (2011).

[44]B. Scharf, A. Matos-Abiague, I. Žutić, and J. Fabian, Phys. Rev. B **85**, 085208 (2012).

[45]*Optical Orientation*, edited by B. P. Zakharchenya and F. Meyer (Elsevier, Amsterdam, 1984).

[46]S. P. Dash, S. Sharma, R. S. Patel, M. P. de Jong, and R. Jansen, Nature (London) **462**, 491 (2009).

[47]S. P. Dash, S. Sharma, J. C. Le Breton, J. Peiro, H. Jaffrès, J.-M. George, A. Lemaître, and R. Jansen, Phys. Rev. B **84**, 054410 (2011).

[48]U. Sivan and Y. Imry, Phys. Rev. B **33**, 551 (1986).

[49]S. O. Valenzuela, D. J. Monsma, C. M. Marcus, V. Narayanamurti, and M. Tinkham, Phys. Rev. Lett. **94**, 196601 (2005).

[50]B. G. Park, T. Banerjee, J. C. Lodder, and R. Jansen, Phys. Rev. Lett. **99**, 217206 (2007).

[51]C. R. Leavens and G. C. Aers, Solid State Commun. **61**, 289 (1987).

[52]J. Marschall and A. Majumdar, J. Appl. Phys. **74**, 4000 (1993).