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ACCEPTED MANUSCRIPT

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Spin caloric transport from density-functional theory

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Abstract. *Spin caloric transport* refers to the coupling of heat with spin transport. Its applications primarily concern the generation of spin currents and control of magnetisation by temperature gradients for information technology, known by the synonym *spin caloritronics*. Within the framework of *ab-initio* theory, new tools are being developed to provide an additional understanding of these phenomena in realistic materials, accounting for the complexity of the electronic structure without adjustable parameters. Here we review this progress, summarising the principles of the density-functional-based approaches in the field and presenting a number of application highlights. Our discussion includes the three most frequently employed approaches to the problem, namely the Kubo, Boltzmann, and Landauer-Büttiker methods. These are showcased in specific examples that span, on the one hand, a wide range of materials, such as bulk metallic alloys, nano-structured metallic and tunnel junctions, or magnetic overlayers on heavy metals, and, on the other hand, a wide range of effects, such as the spin-Seebeck, magneto-Seebeck, and spin-Nernst effects, spin disorder, and the thermal spin-transfer and thermal spin-orbit torques.

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1. Introduction

The field of spin caloric transport bears a close analogy to the field of thermoelectrics. It is standard textbook knowledge [1] that charge and heat currents, $\mathcal{J}$ and $\mathcal{Q}$, are coupled to electric fields $\mathbf{E}$ and temperature gradients ∇T , including cross terms. Considering, in addition, the spin degree of freedom of the travelling electrons, one has a coupling between charge, spin, and temperature gradients, that is the central point of spin-caloric transport. One thus obtains the generalised linear-response equations

$$\mathcal{J} = -\mathcal{L}^{cc}(\nabla\mu + e\mathbf{E}) - \mathcal{L}^{cs}\nabla\mu^s - \mathcal{L}^{cq}\nabla T/T \quad (1)$$

$$\mathcal{J}^s = -\mathcal{L}^{sc}(\nabla\mu + e\mathbf{E}) - \mathcal{L}^{ss}\nabla\mu^s - \mathcal{L}^{sq}\nabla T/T \quad (2)$$

$$\mathcal{Q} = -\mathcal{L}^{qc}(\nabla\mu + e\mathbf{E}) - \mathcal{L}^{qs}\nabla\mu^s - \mathcal{L}^{qq}\nabla T/T \quad (3)$$

Here, $e = |e|$ is the absolute value of the electron charge, μ is the electronic chemical potential, and the quantity $\nabla\mu^e = \nabla\mu + e\mathbf{E}$ is the gradient of the electrochemical potential μ^e . We also have the spin chemical potential, μ^s , corresponding to electron spin accumulation, which is defined as the difference in the chemical potential of the two spin directions, $\mu^s = \mu_{\uparrow} - \mu_{\downarrow}$. Furthermore, $\mathcal{J}^s$ is the spin current, and $\mathcal{L}^{ij}$ are linear coupling coefficients, with the indices i, j taking values in the set $\{c, s, q\}$ that stand for charge, spin, and heat, respectively.

Perhaps the most important spin caloric transport effect is the generation of spin currents by means of thermal gradients, inspiring applications in spintronics, with the scope of developing novel concepts in spin-based information technology [2, 3]. This direction has been termed *spin caloritronics* [2]. Historically, the drive for applications probably even preceded the more fundamental research, so it is fair to say that the field of spin caloric transport was born from spin caloritronics.

The conceptual advances [4] in theory and experiment in spin caloric transport called for materials-specific *ab-initio* calculations, as they are provided by density functional theory (DFT). Mature DFT methods already existed in transport theory, however, the challenging new field demanded further conceptual progress and development of computational tools. As Eqs. (1-3) involve both longitudinal and transversal transport with respect to the applied fields, the area has greatly benefited from the parallel increasing interest and progress in the field of transverse electronic transport in general (especially the anomalous and spin Hall effects).

Thus, modern DFT methods capture longitudinal phenomena, comprising the spin Seebeck, the magneto-Seebeck, and the tunneling magneto-Seebeck effects, as well as transverse phenomena, including the spin and anomalous Nernst effects. These are spin caloric analoga of the standard Seebeck effect, the giant and tunneling magnetoresistance, the spin Hall and the anomalous Hall effects, respectively. Methods were also developed to capture very important secondary effects of spin-caloric transport, with the objective of switching the magnetization direction in magnetic systems. These are the thermal spin torque and the thermal spin-orbit torque as analoga to the longitudinal spin transfer torque and the transverse spin-orbit torque, respectively.

It should be noted that DFT transport methods capture principally the electronic part of the thermal and spin transport. However, just as there is an important phononic part in thermal transport, there also is an important magnonic part in spin transport [5, 6, 7] (dominant in magnetic insulators), as there is a magnonic part in thermal transport [6] and a phononic part in spin transport through a phonon drag mechanism [8]. These important effects are not included in the DFT calculations discussed in the present article, and require additional modelling. Within the limits of electron-mediated transport, we may distinguish three main approaches that are frequently used in the linear-response regime, different in their conceptual modelling of transport and in the physical systems that they target: the Kubo, the Boltzmann, and the Landauer-Büttiker approach.

The purpose of this Topical Review is to accentuate the important on-going DFT-based progress in this rapidly advancing topic of spin caloric transport, by highlighting a number of specific examples that span a wide range of methods and applications by different research groups. The basic principles and the range of applicability of the main approaches are outlined in Sec. 2. Then, sections devoted on applications follow, with each subject introduced individually. Sec. 3 discusses symmetry aspects of the linear response coefficients based on the Kubo formalism. In Sec. 4, the Berry phase approach is applied to the effect of the (transverse) thermal spin-orbit torque. Sec. 5 concerns the semiclassical Boltzmann formalism and its application to calculations in dilute alloys. Applications of the Landauer-Büttiker formalism are presented in the following sections. Sec. 6 investigates spin caloric trans-

port in nanostructured Heusler alloys. Sec. 7 presents the theory of the tunneling magneto-Seebeck effect and the thermal spin-transfer torque. Finally, Sec. 8 inspects the effect of spin disorder at elevated temperatures on spin-caloric transport properties. A brief summary and outlook is presented in Sec. 9.

In addition to the aforementioned highlights, we note that density-functional methods are being increasingly applied to spin-caloric transport in molecular-based nano-contacts (see, e.g., refs. [9, 10]), where the richness of combinations, featuring magnetic leads, magnetic molecules, or both, opens a number possibilities in spin-transport control [11].

2. General remarks on the theoretical approaches. The role of Green functions

All approaches to the electronic contribution to spin-caloric or thermoelectric transport target primarily the calculation of the energy-dependent charge conductivity $\sigma_c(E)$ or spin conductivity $\sigma_s(E)$. Conventional wisdom requires $\sigma_{c/s}(E)$ to be second-rank, with its components referring to the applied field and the resulting current direction. In the case of spin transport, however, one must also consider the direction of polarisation of the spin quantities $\nabla\mu^s$ and $\mathcal{J}^s$, which results in higher-rank tensors.

The conductivity tensor has two contributions: one extrinsic, the other intrinsic. The extrinsic contribution depends on the scattering details of the electrons and is confined to the vicinity of the Fermi level. It partakes both in longitudinal as well as in transverse transport. The intrinsic contribution, on the other hand, is principally important for transverse transport and became widely known through the theory of the anomalous and spin Hall effects. It arises as the electrons, accelerated by the electric field, are promoted to different bands and thus sample the relative phases of different Bloch wavefunctions.

Having calculated $\sigma_{c/s}(E)$ in a range of energies around the Fermi level E_F , these quantities are convoluted with the Fermi function $f^0(E, \mu, T)$ in the standard formulas

$$L_n^{c/s} = -\frac{1}{e} \int dE \sigma_{c/s}(E) \left[-\frac{\partial f^0(E, \mu, T)}{\partial E} \right] (E - \mu)^n \quad (4)$$

with $n = 0, 1$, or 2 . One power of $(E - \mu)$ derives from the thermal current (an electron at energy E carries an amount of heat equal to $E - \mu$), and one derives from the spatial derivative of the Fermi function accompanying ∇T . Finally, it is straightforward to identify the linear coefficients $\mathcal{L}^{ij}$ of Eqs. (1-3) with the appropriate $L_n^{c/s}$, in analogy to the standard theory of thermoelectricity: $\mathcal{L}^{cc} = L_0^c$, $\mathcal{L}^{cq} = L_1^c$, $\mathcal{L}^{qq} = L_2^c$, $\mathcal{L}^{sc} = L_0^s$, etc. In the limit $T \rightarrow 0$, the derivative

$-\partial f^0(E, \mu, T)/\partial E$ approaches $\delta(E - E_F)$, and the conduction is pinned on the Fermi level.

As an example, we discuss the Seebeck and spin-Seebeck coefficients. The Seebeck coefficient quantifies the electric field (actually the electrochemical potential gradient in absence of externally applied electric fields) that is induced as a response to a thermal gradient under open circuit conditions, $\nabla\mu^e/e = S\nabla T$, with

$$S = -\frac{1}{eT} \frac{L_1^c}{L_0^c}. \quad (5)$$

In analogy, in spin caloric transport, one defines the spin-Seebeck coefficient, that quantifies the spin accumulation gradient as a response to a thermal gradient, $\nabla\mu^s/e = S^{\text{spin}}\nabla T$, with

$$S^{\text{spin}} = -\frac{1}{eT} \frac{L_1^s}{L_0^s}. \quad (6)$$

There is, however, a complication. Since Eq. (5) reflects the electrochemical potential that is formed under open circuit conditions ($\mathcal{J} = 0$), it describes the effect of charge accumulation on the two sides of the open circuit. In analogy, Eq. (6) should reflect the gradient in spin chemical potential that is formed under the analogous conditions ($\mathcal{J}^s = 0$). However, this interpretation may sometimes lead to confusion, because the electron spin is not a conserved quantity. Spin currents are possible under open circuit conditions, with spins sourced by the fields and drained by spin-flip processes, i.e., interactions of the electron spin with the lattice mediated through impurities, localised magnetic moments, nuclear spins, etc., that may depend to a very large extent on the specific experimental situation and the fine details of the sample. ‡ With respect to this possible ambiguity, we accept Eq. (6) as a definition of the (calculated) spin Seebeck coefficient, leaving the question of its correct interpretation open to each particular application.

In electron transport phenomena, one must consider the electron scattering in the sample. In the cases where the electron phase relaxation length is shorter than the sample size (diffusive transport), the main sources of electron scattering are defects (or disorder in general), atomic vibrations, magnetic fluctuations, or electron-electron interactions. In the cases that the electron phase relaxation length is comparable to, or longer than, the sample size (ballistic transport), e.g. in tunnel junctions, an additional and usually dominant source of scattering is the interface of the sample to the leads.

A DFT error that can affect tunneling transport calculations is the underestimation of the band gap in the insulating spacer. If the Fermi energy falls

‡ Spin currents are possible even without external fields, but here we do not address these effects.

in the middle of the gap, this error may lead to an underestimation of the tunneling resistance. In this case, however, the relative difference of resistance between two states (magnetoresistance or, in analogy, magneto-Seebeck coefficient, see e.g. Sec. 7) is less prone to the error.

Within the DFT methods, the sources of scattering are usually treated by approximating the scattering potential as being time-independent. While this is obvious for impurities or interfaces, it requires adopting the adiabatic approximation for atomic vibrations and magnetic fluctuations. In both cases, this approximation rests on time-scale arguments: the motion of atoms and the precession of atomic magnetic moments is much slower than the motion of electrons [12, 13]. On the other hand, the electron-electron interaction is only treated on a mean-field level by using the potential produced by the time-independent density. At the same time, the tacit approximation is made that the current matrix elements are calculated between Kohn-Sham wavefunctions and that only elastic scattering is considered. These assumptions are reasonable as long as DFT provides a good description of the ground-state electronic structure and as long as one is close to a Fermi-liquid behaviour, which is expected close to the Fermi level in the linear response regime. Comparison with experiment has shown the success of these approximations [14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24].

Given the above approximations, we may summarise the three principal approaches as follows.

- The *Kubo approach* is the most general among the approaches in the diffusive regime. It can treat any degree of disorder in a supercell mode, but it has primarily been applied in connection with the coherent potential approximation (CPA) to chemical disorder and atomic vibrations. It addresses both the extrinsic and intrinsic contributions to transverse transport. The central quantity is the current-current correlation function. A special case of the Kubo approach is the *Berry phase approach* for transverse transport, that focuses on the intrinsic contribution of the bands.
- The *Boltzmann approach*, semi-classical in its modelling, is appropriate for dilute alloys, or more generally translationally periodic systems with a low degree of disorder. Concerning the transverse transport phenomena, it can treat the extrinsic contribution only, not the intrinsic one. The central quantity is the mean free path, which should be longer than the assumed Bloch wavepacket spread, but shorter than the sample size, for the approach to be formally applicable. It has limited applicability compared

to the Kubo approach, but on the other hand it is intuitively more transparent because it reduces the problem to two separate constituents, i.e., the band structure part and the scattering part.

- The *Landauer-Büttiker approach* models systems in a junction geometry, namely a region where scattering takes place, in contact with perfectly conducting leads. Even if it was initially designed for ballistic or tunneling transport, it has been also applied with success to diffusive problems by considering a large volume, where scattering takes place because of disorder, between perfectly conducting leads. The Landauer-Büttiker approach requires some obvious adjustments of Eqs. (1-4): the electric field is replaced by a bias voltage across the junction, the current density by the current, the conductivity by the conductance. It treats longitudinal as well as transverse [25] transport phenomena. Some groups have extended it to the non-linear response regime by applying the non-equilibrium Green function technique, particularly suited for finite-bias tunneling problems.

The aforementioned approaches require in most cases a Green function formalism, since one deals with response functions. In the Kubo approach, the conductivity tensor is calculated from a product of Green functions of the form $\sigma \sim \langle \hat{J} \mathcal{G} \hat{J} \mathcal{G} \rangle$, where $\mathcal{G}$ is the Green function, $\hat{J}$ is the current operator and $\langle \dots \rangle$ stands for an average over the disorder configurations. The conductivity obtains a “Fermi-surface contribution” (constant energy part) that suffices for the longitudinal transport and a “Fermi-sea contribution” that is necessary for the description of the transverse transport, resulting in the Kubo-Bastin formula (see Sec. 3 for more details). In the cases where the CPA is used for a description of the disorder, vertex corrections are necessary (formally at least), because the CPA without vertex corrections only gives $\langle \mathcal{G} \rangle$, i.e., averages the Green function, not the product of Green functions.

From the Kubo formula, and under some simplifying assumptions, one arrives at a geometrical (Berry) phase formulation for the transverse conductivity that accounts for the anomalous velocity. The calculation involves the periodic part of the Bloch wavefunctions $u_{\mathbf{k}}(\mathbf{r})$ and the Berry curvature, which is integrated over all occupied states (Fermi sea). Numerically, this is greatly simplified by a transformation to a basis of Wannier functions.

The simplifying physical assumption of the Berry phase formulation is to neglect the extrinsic contribution of the scattering; rather, it is assumed that the transverse current comes only from the deformation of the initial Bloch states of the band at energy $E_{n\mathbf{k}}$ by the external electric field, which

is realized by interband transitions to other bands $E_{n'\mathbf{k}}$ that are folded down to the initial band by using the Berry phase technique. The Berry phase formulation is thus applicable either at elevated temperatures (where atomic vibrations dominate the scattering), or to moderately dirty samples, under the additional assumption that, under disorder, the crystal momentum is still a reasonably defined quantity (“sharp bands”) and that the intrinsic contribution outweighs the extrinsic one. To some extent, the degree of disorder and temperature-induced atomic vibrations are approximated by introducing using a single parameter, the relaxation time τ .

In the Boltzmann approach, the Green functions are used to calculate the scattering amplitude (t -matrix) $T_{\mathbf{k}\mathbf{k}'} = (\Psi_{\mathbf{k}}, \Delta V \Psi_{\mathbf{k}'}^{\text{sc}})$ between initial scattered states $\Psi_{\mathbf{k}'}^{\text{sc}}$ and final Bloch states $\Psi_{\mathbf{k}}$, where ΔV is the scattering potential of the defects. The t -matrix is connected to the Green functions of the crystal, $\mathcal{G}_0$, and of the defective system, $\mathcal{G}_{\text{def}}$, through the equations $T = \Delta V + \Delta V \mathcal{G}_0 T$ and $T = \Delta V + \Delta V \mathcal{G}_{\text{def}} \Delta V$, respectively. Given the t -matrix, the scattering rate is calculated by the Golden Rule: $P_{\mathbf{k}\mathbf{k}'} = \frac{2\pi}{\hbar} |T_{\mathbf{k}\mathbf{k}'}|^2 \delta(E_{\mathbf{k}} - E_{\mathbf{k}'})$. In the presence of many impurities, the multiple-scattering interference is usually neglected, and it is assumed that the rate $P_{\mathbf{k}\mathbf{k}'}$ scales linearly with the impurity concentration. The rate enters the scattering expression of the Boltzmann equation. Even though the Boltzmann formalism accounts for the scattering in detail, it misses the intrinsic contribution to the transverse phenomena. In this respect, the Berry phase formalism and the Boltzmann formalism are complementary to each other.

In the Landauer-Büttiker approach, a junction geometry is set up with two ideally conducting leads sandwiching the scattering region. For the description of the half-infinite leads, the associated self-energy is needed, which is closely related to the Green function. Also, the matching of the scattering region to the leads is conveniently formulated in terms of the Green function. Finally, one must calculate the transmission coefficient between channels of the left and right leads, which may also be conveniently done by calculating the Green function elements between the left and right lead, convoluted with the current operator. In effect, the Landauer-Büttiker formalism is a spatially integrated form of the Kubo formalism. It can be used to calculate not only longitudinal, but also transverse transport, including intrinsic contributions, as has been demonstrated in [25].

3. Spin-dependent thermo-galvanic effects in disordered alloys

3.1. Kubo's linear response formalism

The following gives an account of various recent extensions and applications of Kubo's linear response formalism to deal with spin-dependent transport in response to a gradient of the electrochemical potential $\nabla\mu^e = \nabla\mu + e\mathbf{E}$ and a gradient of the temperature ∇T , where μ is the chemical potential, $e = |e|$ the elementary charge and $\mathbf{E}$ the electric field.

Spin transport induced by these perturbations can be discussed in full analogy to the treatment of charge transport based on Kubo's formalism [26, 27]. Assuming a constant chemical potential and considering a spin-polarised current $\mathcal{J}^s$ due to the simultaneous presence of $\mathbf{E}$ as well as ∇T , one may write

$$\mathcal{J}^s = -e\mathcal{L}^{\text{sc}}\mathbf{E} - \mathcal{L}^{\text{sq}}\nabla T/T, \quad (7)$$

with the corresponding third rank response tensors $\mathcal{L}^{\text{sc}}$ and $\mathcal{L}^{\text{sq}}$. The $\mu\nu\xi$ -component of $\mathcal{L}^{\text{sc}}$ can be expressed in terms of the corresponding element $\sigma_{\mu\nu}^{\text{sc},\xi}(E)$ of the energy- (E) -dependent spin conductivity tensor,

$$\mathcal{L}_{\mu\nu}^{\text{sc},\xi}(T) = -\frac{1}{e} \int dE \sigma_{\mu\nu}^{\text{sc},\xi}(E) D(E, E_F, T), \quad (8)$$

where the spin polarisation is along ξ , the spin current along μ , the electric field along ν and $D(E, E_F, T) = -\partial f^0(E, E_F, T)/\partial E$ with $f^0(E, E_F, T)$ the Fermi function and E_F the Fermi energy (μ of the electrons at $T = 0$ K).

In analogy to the charge transport [28, 29], the thermally-induced spin transport coefficient $\mathcal{L}_{\mu\nu}^{\text{sq},\xi}(T)$ can be expressed in terms $\sigma_{\mu\nu}^{\text{sc},\xi}(E)$ as

$$\mathcal{L}_{\mu\nu}^{\text{sq},\xi}(T) = -\frac{1}{e} \int dE \sigma_{\mu\nu}^{\text{sc},\xi}(E) D(E, E_F, T) (E - E_F). \quad (9)$$

Kubo's linear response formalism provides a very powerful basis to calculate the spin conductivity $\sigma_{\mu\nu}^{\text{sc},\xi}(E)$ and this way $\mathcal{L}_{\mu\nu}^{\text{sc},\xi}(T)$ and $\mathcal{L}_{\mu\nu}^{\text{sq},\xi}(T)$ via Eqs. (7) and (9), respectively. Considering the rather general case of a response observable represented by an arbitrary operator product of the form $(\hat{B}_\mu \hat{C}_\xi)$ and a perturbation by the operator $\hat{A}_\nu$, with $\hat{A}_\nu$, $\hat{B}_\mu$, and $\hat{C}_\xi$ the Cartesian components of vector operators, one has for the corresponding frequency- (ω) -dependent response function

$$\tau_{(\hat{B}_\mu \hat{C}_\xi) \hat{A}_\nu}(\omega, \mathbf{H}) = \int_0^\infty dt e^{-i\omega t} \int_0^\beta d\lambda \text{Tr} \left(\rho(\mathbf{H}) \hat{A}_\nu \hat{B}_\mu(t + i\hbar\lambda; \mathbf{H}) \hat{C}_\xi(t + i\hbar\lambda; \mathbf{H}) \right) \quad (10)$$

where ρ is the density operator, $\beta = 1/k_B T$ with k_B the Boltzmann constant, and $\mathbf{H}$ is a magnetic field [30].

Describing the electronic structure within the framework of density functional theory (DFT) or another adequate single-particle picture Eq. (10) simplifies to the so-called Kubo-Bastin formula [31] that uses a representation of the electronic structure by means of the retarded and advanced Green function operators, $\hat{G}^+$ and $\hat{G}^-$, respectively. Dealing with the response to a static electric field only the case $\omega = 0$ has to be considered with the perturbation represented by the electric current density operator $\hat{A}_\nu = \hat{j}_\nu$. For the electrical conductivity one has for the observable $\hat{C}_i = 1$ and $\hat{B}_\mu = \hat{j}_\mu$, while for the spin conductivity one has to replace $(\hat{B}_\mu \hat{C}_\xi)$ by an appropriate spin current density operator $\hat{J}_\mu^\xi$ leading to the corresponding Kubo-Bastin formula for $\sigma_{\mu\nu}^\xi$ [32]:

$$\sigma_{\mu\nu}^\xi = \frac{\hbar}{4\pi\Omega} \text{Tr} \quad (11)$$

$$\left[\left\langle \hat{J}_\mu^\xi (\hat{G}^+ - \hat{G}^-) \hat{j}_\nu \hat{G}^- - \hat{J}_\mu^\xi \hat{G}^+ \hat{j}_\nu (\hat{G}^+ - \hat{G}^-) \right\rangle \right.$$

$$+ \int_{-\infty}^{E_F} dE \left\langle \hat{J}_\mu^\xi \hat{G}^+ \hat{j}_\nu \frac{d\hat{G}^+}{dE} - \hat{J}_\mu^\xi \frac{d\hat{G}^+}{dE} \hat{j}_\nu \hat{G}^+ \right.$$

$$\left. - \left(\hat{J}_\mu^\xi \hat{G}^- \hat{j}_\nu \frac{d\hat{G}^-}{dE} - \hat{J}_\mu^\xi \frac{d\hat{G}^-}{dE} \hat{j}_\nu \hat{G}^- \right) \right\rangle.$$

As the calculation of spin transport coefficients requires to account for spin-orbit coupling within the underlying electronic structure calculations a fully relativistic approach based on the Dirac formalism [33] has been adopted within the present work. The corresponding real space representation of operators j_ν and $\hat{J}_\mu^\xi$ have been discussed in detail in Refs. [34, 19, 32]. For the representation of the Green function operators $\hat{G}^+$ and $\hat{G}^-$ the multiple scattering or KKR (Korringa-Kohn-Rostoker) band structure method has been used as described elsewhere [35, 33]. Combining this approach with the Coherent Potential Approximation (CPA) allows to account in a coherent way for chemical disorder in alloys as it was demonstrated for the non-relativistic case by Butler [36]. The corresponding configurational average, indicated by the brackets $\langle \dots \rangle$ in Eq. (11), accounts in particular for the so-called vertex corrections, that are of central importance when dealing with transverse charge or spin transport [37, 38].

Eq. (10) with the restriction $\hat{C}_i = 1$ was used by Kleiner [30] to derive for all magnetic space groups the shape of the tensors describing charge and heat transport in response to an electric field or temperature gradient. His scheme was extended by us [39] to the more general form given in Eq. (10) by working out the transformation behaviour of the response function $\tau_{(\hat{B}_\mu \hat{C}_\xi) \hat{A}_\nu}(\omega, \mathbf{H})$ under a symmetry operation. Considering for example the unitary operation u one

finds [39]:

$$\tau_{(\hat{B}_\mu \hat{C}_\xi) \hat{A}_\nu}(\omega, \mathbf{H}) = \sum_{lmn} \tau_{(\hat{B}_m \hat{C}_n) \hat{A}_l}(\omega, \mathbf{H}_u) \quad (12)$$

$$D^{(\hat{A})}(u)_{lv} D^{(\hat{B})}(u)_{m\mu} D^{(\hat{C})}(u)_{n\xi},$$

where the Wigner D-matrices $D^{(\hat{X})}(u)$ represent the transformation behaviour of an operator $\hat{X}$ under operation u . A similar relation holds for an anti-unitary operation a that involves time reversal [39]. Collecting all restrictions imposed by these symmetry relations leads to corresponding restrictions for the shape of the considered response tensor $\tau_{(\hat{B}_\mu \hat{C}_\xi) \hat{A}_\nu}(\omega, \mathbf{H})$.

Applying the above to the inverse thermoelectric or Peltier ($\underline{\alpha}'$), electrical ($\underline{\sigma}$) or spin ($\underline{\sigma}^k$) conductivity tensors, the transformation behaviour of the involved operators allows restricting only to the 32 possible magnetic Laue groups instead of all 1651 magnetic space or 122 magnetic point groups. Table 1 gives as an example the corresponding tensor shapes found on the basis of Eq. (12) for the magnetic Laue groups $m\bar{3}m1'$, $4/m\bar{m}'m'$, $4/m1'$, and $2/m1'$ [39]. Concerning the complete set of results [39], one first of all has to note that all tensor shapes given by Kleiner [30], who used a less common definition of the (magnetic) Laue group, have been reproduced. Comparing the shape of the tensors $\underline{\alpha}'$ and $\underline{\sigma}$ given in Table 1 for the magnetic Laue groups $4/m1'$, and $2/m1'$, one finds that these are not identical. Obviously, this is in contradiction to the common practise to use the Mott formula [40] or its generalisation to tensors and finite temperatures, that give –in analogy to Eq. (8) for $\mathcal{L}_{\mu\nu}^{\text{sq},\xi}$ – the transport coefficient $\mathcal{L}_{\mu\nu}^{\text{eq}}$ in terms of the electrical conductivity $\sigma_{\mu\nu}^{\text{cc}}(E)$, also for the off-diagonal elements of the thermoelectric tensor $\underline{\alpha}$, the Onsager reciprocal of $\underline{\alpha}'$. This implies that the use of Mott-like formulae in this context has to be reconsidered. For spin transport due to an electric field or temperature gradient the situation is different. In this case the symmetry analysis shows that the tensors representing the response to an electric field and to a temperature gradient have to have the same shape. A detailed discussion of the appearance of longitudinal spin-polarised transport in nonmagnetic systems suggested by the very last columns in the last two lines of Table 1 can be found in Ref. [41].

3.2. Anisotropy of the Seebeck effect and anomalous Nernst effect in ferromagnetic alloys

The first two examples given in Table 1 clearly show that spin polarisation of a material together with spin-orbit coupling (SOC) gives rise to pronounced qualitative changes of the transport properties of a magnetic material when compared to its non-magnetic reference state.

magnetic Laue group	$\underline{\alpha}'$	$\underline{\sigma}$	$\underline{\sigma}^x$	$\underline{\sigma}^y$	$\underline{\sigma}^z$
$m\bar{3}m1'$ (fcc-Pt)	$\begin{pmatrix} \alpha_{xx} & 0 & 0 \\ 0 & \alpha_{xx} & 0 \\ 0 & 0 & \alpha_{xx} \end{pmatrix}$	$\begin{pmatrix} \sigma_{xx} & 0 & 0 \\ 0 & \sigma_{xx} & 0 \\ 0 & 0 & \sigma_{xx} \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & \sigma_{yz}^x \\ 0 & -\sigma_{yz}^x & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & -\sigma_{yz}^x \\ 0 & 0 & 0 \\ \sigma_{yz}^x & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & \sigma_{yz}^x & 0 \\ -\sigma_{yz}^x & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$
$4/m\bar{m}'m'$ (fcc-Co _x Pd _{1-x})	$\begin{pmatrix} \alpha_{xx} & \alpha_{xy} & 0 \\ -\alpha_{xy} & \alpha_{xx} & 0 \\ 0 & 0 & \alpha_{zz} \end{pmatrix}$	$\begin{pmatrix} \sigma_{xx} & \sigma_{xy} & 0 \\ -\sigma_{xy} & \sigma_{xx} & 0 \\ 0 & 0 & \sigma_{zz} \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & \sigma_{xz}^x \\ 0 & 0 & \sigma_{yz}^x \\ \sigma_{zx}^x & \sigma_{zy}^x & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & -\sigma_{xz}^x \\ 0 & 0 & \sigma_{xz}^x \\ -\sigma_{zy}^x & \sigma_{zx}^x & 0 \end{pmatrix}$	$\begin{pmatrix} \sigma_{xx}^z & \sigma_{xy}^z & 0 \\ -\sigma_{xy}^z & \sigma_{xx}^z & 0 \\ 0 & 0 & \sigma_{zz}^z \end{pmatrix}$
$4/m1'$ (Au ₄ Sc)	$\begin{pmatrix} \alpha_{xx} - \alpha_{xy} & 0 \\ \alpha_{xy} & \alpha_{xx} & 0 \\ 0 & 0 & \alpha_{zz} \end{pmatrix}$	$\begin{pmatrix} \sigma_{xx} & 0 & 0 \\ 0 & \sigma_{xx} & 0 \\ 0 & 0 & \sigma_{zz} \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & \sigma_{xz}^x \\ 0 & 0 & \sigma_{yz}^x \\ \sigma_{zx}^x & \sigma_{zy}^x & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & -\sigma_{xz}^x \\ 0 & 0 & \sigma_{xz}^x \\ -\sigma_{zy}^x & \sigma_{zx}^x & 0 \end{pmatrix}$	$\begin{pmatrix} \sigma_{xx}^z & \sigma_{xy}^z & 0 \\ -\sigma_{xy}^z & \sigma_{xx}^z & 0 \\ 0 & 0 & \sigma_{zz}^z \end{pmatrix}$
$2/m1'$ (Pt ₃ Ge)	$\begin{pmatrix} \alpha_{xx} & \alpha_{yx} & 0 \\ \alpha_{xy} & \alpha_{yy} & 0 \\ 0 & 0 & \alpha_{zz} \end{pmatrix}$	$\begin{pmatrix} \sigma_{xx} & \sigma_{xy} & 0 \\ \sigma_{xy} & \sigma_{yy} & 0 \\ 0 & 0 & \sigma_{zz} \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & \sigma_{xz}^x \\ 0 & 0 & \sigma_{yz}^x \\ \sigma_{zx}^x & \sigma_{zy}^x & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & \sigma_{xz}^y \\ 0 & 0 & \sigma_{yz}^y \\ \sigma_{zx}^y & \sigma_{zy}^y & 0 \end{pmatrix}$	$\begin{pmatrix} \sigma_{xx}^z & \sigma_{xy}^z & 0 \\ \sigma_{yx}^z & \sigma_{yy}^z & 0 \\ 0 & 0 & \sigma_{zz}^z \end{pmatrix}$

Table 1. Inverse thermoelectrical or Peltier ($\underline{\alpha}'$), electrical ($\underline{\sigma}$) and spin ($\underline{\sigma}^k$) conductivity tensor shapes for four magnetic Laue groups [39]. Below each group symbol an example for a corresponding material is given in parentheses.

Considering a cubic metal with its magnetisation along $\hat{z}$, the shape of the conductivity tensor $\underline{\sigma}$ implies the occurrence of the anisotropic magneto-resistance (AMR), i.e., the corresponding resistivity depends on the orientation of the electric field with respect to the magnetisation. The AMR is usually expressed in terms of the ratio $(\rho_{zz} - \rho_{xx})/\rho_{iso}$, where ρ_{iso} is the average over the diagonal elements of the resistivity tensor $\underline{\rho} = (\underline{\sigma})^{-1}$. As implied by the generalised Mott formula, i.e., the equivalent of Eqs. (8) and (9) for charge transport, this anisotropy will be carried over to thermoelectric transport. Imposing for example open-circuit conditions for the charge current an internal electric field builds up according to $\mathbf{E} = \mathbf{S} \nabla T$, where $\mathbf{S} = -\underline{\sigma}^{-1} \underline{\alpha}$ is the thermo(magneto)electric tensor. The anisotropy reflected by the shape of $\mathbf{S}$ implies that the Seebeck coefficients S_{ii} for a temperature gradient perpendicular (xx) and parallel (zz) to the magnetisation are different with the corresponding anisotropy of the Seebeck effect (ASE) expressed by the ratio $(S_{xx} - S_{zz})/S_{iso}$.

The relation of the AMR and ASE as a function of the concentration has been investigated in detail for the transition metal alloy Co_xPd_{1-x} [21]. Fully in line with experiment [42] it is found that the AMR ratio rises steeply for an increasing Co content up to about $x = 0.25$. While the experimental ratio is nearly constant, the theoretical values drop again if x is further increased. This pronounced concentration dependence of the AMR ratio could in part be related to specific features of the electronic structure and their variation with composition. Kudrnovský *et al.* could later show that the poor agreement for intermediate concentrations can be traced back to partial $L1_0$ ordering [43]. For the ASE ratio a rather

sharp maximum at around $x = 0.2$ is observed. Its magnitude of nearly 0.2 is rather large when compared with available experimental values found for other systems [44, 45, 46, 47]. Although AMR and ASE show a maximum at approximately the same composition x , their overall variation is quite different and no one-to-one relation between these quantities is found. This is in fact not to be expected, as the integral kernels in the expressions for $\mathcal{L}_{\mu\nu}^{cc}$ and $\mathcal{L}_{\mu\nu}^{cq}$, corresponding to Eqs. (8) and (9), respectively, differ by the factor $(E - E_F)$.

As reflected by the off-diagonal elements of the conductivity tensor $\underline{\sigma}$ another consequence of SOC for magnetic solids is the phenomenon of transverse charge transport, i.e., the occurrence of the anomalous Hall effect (AHE). Fig. 1 (top) shows corresponding results for the anomalous Hall conductivity σ_{xy} for $T = 0$ K of Co_xPd_{1-x} in comparison with experimental data [48] that are reproduced in a very satisfying way. Calculating σ_{xy} with (VC) and without (NV) the so-called vertex corrections [36, 49] allow one to identify the extrinsic contributions to σ_{xy} due to the skew scattering and side jump mechanisms [50, 37]. As one can see, these are quite pronounced in the Pd- as well as Co-rich regimes, however, having different sign at the two sides of the alloy system. This situation is obviously very much the same as found for the spin Hall effect in non-magnetic transition-metal alloys [38].

As for the longitudinal transport the AHE has a thermoelectric analog, the anomalous Nernst effect (ANE). The corresponding anomalous Nernst conductivity (ANC) α_{xy} for $T \rightarrow 0$ of Co_xPd_{1-x} (see Fig. 1, bottom) shows a very prominent maximum around $x = 0.2$. In contrast to the AHE, the impact of the vertex corrections is rather weak, in particular

Figure 1. (Color online) Top: calculated AHC (VC, full squares) together with its intrinsic contribution (NV, open squares) in comparison to low temperature experimental data (circles) [48]. Bottom: calculated ANC α_{xy} (VC, full triangles) together with its intrinsic contribution (NV, open triangles), in the athermal limit. Reproduced from Ref. [21].

for large x , as the difference of the results obtained with (VC) and without (NV) them demonstrates. This implies that the intrinsic contribution to α_{xy} dominates throughout almost the entire concentration regime considered. For the same reason given in case of the AMR and ASE, there is obviously no correspondence between $\alpha_{xy}(x)$ and $\sigma_{xy}(x)$.

3.3. Spin Nernst effect in diluted alloys – Kubo vs. Boltzmann formalism

Turning to explicitly spin-dependent transport, the spin Hall effect represented for the case of a cubic nonmagnetic system by the coefficient σ_{yx}^z in the last three columns of the first line in Table 1 also has a thermally driven counterpart, the spin Nernst effect [51, 52]. As discussed above, replacing the electric charge current density operator by the electric heat current density in $\hat{A}_\nu$ of Eq. (10) does not lead to modified symmetry restrictions, accordingly Eq. (9) can be used to calculate the spin Nernst conductivity $\alpha_{\mu\nu}^\xi = -\mathcal{L}_{\mu\nu}^{\text{sq},\xi}/T$.

The first *ab-initio* investigations on the spin Nernst effect were performed by Tauber *et al.* on the basis of the linearized Boltzmann transport equation and using a relativistic spin-projection scheme [53]. Calculations were carried out for the diluted alloys $\text{Cu}_{0.99}\text{M}_{0.01}$ with $\text{M} = \text{Ti}, \text{Au}, \text{and Bi}$ and limited to the skew scattering contribution.

Imposing open-circuit conditions for the charge current, Eq. (7) can be reformulated as

$$\mathcal{J}^s = \mathcal{L}^{\text{sc}}(-e\mathbf{E}) + \mathcal{L}^{\text{sq}}(-\nabla T/T) = \alpha^{\text{scq}} \nabla T, \quad (13)$$

with the third-rank tensor

$$\begin{aligned} \alpha^{\text{scq}} &= -e\mathcal{L}^{\text{sc}}\underline{S} - \mathcal{L}^{\text{sq}}/T \\ &= \mathcal{L}^{\text{sc}}(\mathcal{L}^{\text{cc}})^{-1}\mathcal{L}^{\text{sq}}/T - \mathcal{L}^{\text{sq}}/T. \end{aligned} \quad (14)$$

For the transverse components with respect to the polarisation axis in $\xi = z$ one finds for the magnetic Laue group $m\bar{3}m1'$ the only nonvanishing term

$$\alpha_{yx}^{\text{scq},z} = -e\mathcal{L}_{yx}^{\text{sc},z}S_{xx} - \frac{1}{T}\mathcal{L}_{yx}^{\text{sq},z} = \alpha_{yx}^{\text{sc},z} + \alpha_{yx}^{\text{sq},z}, \quad (15)$$

consisting of an “electrical” contribution $\alpha_{yx}^{\text{sc},z} = \sigma_{yx}^z S_{xx}$ and a “thermal” contribution $\alpha_{yx}^{\text{sq},z}$ from the spin Nernst effect.

A comparison between results based on the Kubo and Boltzmann transport formalisms is shown in Fig. 2 for the total antisymmetric *spin*caloric conductivity $\alpha_{yx}^{\text{scq},z}$ as well as its individual contributions [54]. The

Figure 2. (Color online) Temperature dependence of the total transverse spin caloric conductivity $\alpha_{yx}^{\text{scq},z}$ and its constituents in $\text{Cu}_{0.99}\text{Ti}_{0.01}$, obtained within Kubo and Boltzmann [53] transport theory. Reproduced from Ref. [54].

linear behaviour, relative magnitude and sign of the two terms $\alpha_{yx}^{\text{sc},z}$ and $\alpha_{yx}^{\text{sq},z}$ found in Ref. [53] are reproduced, implying that both are dominated by the skew scattering mechanism, as could indeed be demonstrated (see below). Just as for the Seebeck coefficient, magnitude, sign and temperature dependence of $\alpha_{yx}^{\text{sq},z}$ can be already qualitatively estimated from the energy dependence of the spin Hall conductivity [54].

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3.4. Spin Hall and Nernst effect in concentrated alloys

Unlike the Boltzmann transport equation, Kubo's linear response formalism is not limited in its application to the dilute limit and accounts for all intrinsic and extrinsic scattering mechanisms on the same footing.

The nonmagnetic alloy systems $\text{Au}_x\text{Cu}_{1-x}$ was chosen for a comparative investigation of the concentration dependence of the spin Hall and spin Nernst conductivities and their intrinsic and extrinsic contributions [54]. Figure 3 shows the Mott-like, athermal limits of the electrical and thermal contributions to the total transverse spincaloric conductivity, $\alpha_{yx}^{\text{sc},z}/T$ and $\alpha_{yx}^{\text{sq},z}/T$, respectively. The intrinsic contributions, ob-

Figure 3. (Color online) Components of the total transverse spincaloric conductivity for $T \rightarrow 0$ in $\text{Au}_x\text{Cu}_{1-x}$, excluding and including the vertex corrections. Reproduced from Ref. [54].

tained by ignoring the vertex corrections, are in both cases rather small and vary almost linearly with x . In the case of the electrical contribution this is the combined effect of the increase of σ_{yx}^z due to the increasing average SOC strength and the simultaneous increase of S_{xx} due to an increasingly pronounced slope of $\sigma_{xx}(E)$ in the vicinity of E_F .

Inclusion of the vertex corrections leads to strong apparently diverging extrinsic contributions in the low-concentration regimes, i.e., for x close to 0 or 1, respectively. In case of $\alpha_{yx}^{\text{sc},z}/T$ this can be predominantly ascribed to the spin Hall conductivity, as the impact of the vertex corrections on the Seebeck coefficient is only minor. Making use of the different scaling behavior [55, 56] of the extrinsic contributions to σ_{yx}^z one finds that the side-jump part is, as the intrinsic contribution, quite small and only weakly concentration dependent, but opposite in sign. As a consequence, the skew scattering contribution dominates by far in the low-concentration regimes (see supplemental material of Ref. [54]).

The thermal contribution $\alpha_{yx}^{\text{sq},z}/T$ also shows a

diverging behavior but with opposite sign for $x \rightarrow 0$ and $x \rightarrow 1$. This clearly demonstrates once again that there is no trivial correspondence between the spin Hall conductivity (SHC) σ_{yx}^z and the spin Nernst conductivity (SNC) $\alpha_{yx}^{\text{sq},z}/T$ as discussed above for the charge transport coefficients. Making again use of the connection between the vertex corrections and the extrinsic contributions to the spin conductivity, one finds—similarly to the SHC—only a small and linearly varying intrinsic contributions to the SNC. The extrinsic contributions are again prevailing in the dilute-concentration regimes of $\text{Au}_x\text{Cu}_{1-x}$, as shown in Fig. 3.

Figure 4. (Color online) Decomposition of the spin Nernst conductivity on the Cu-rich side of $\text{Au}_x\text{Cu}_{1-x}$ into intrinsic and extrinsic (skew scattering and side-jump) contributions.

Based on a very similar scaling behavior of the extrinsic contributions due to the skew scattering and side-jump mechanisms w.r.t. to the longitudinal thermoelectrical conductivity a corresponding decomposition has been made at the Cu-rich side of the concentration range for the SNC. Fig. 4 shows that the side-jump contribution is in the same order of magnitude as the intrinsic one and also varies only slightly with concentration. The skew scattering contribution, on the other hand, gives again rise to the diverging behavior when approaching the dilute limit. Taken together, it could be shown that the relative importance and concentration dependence of the different scattering mechanisms of the SNC in diluted transition metal alloys is very similar to the behaviour known from the SHC.

3.5. Spin Nernst Magnetothermopower

Applying a longitudinal temperature gradient to a thin platinum film deposited on top of the insulating ferrimagnet yttrium iron garnet (YIG) and modulating the orientation of the magnetisation by means of an external magnetic field, the spin Nernst effect could

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be recently verified experimentally [52]. Depending on the relative orientation of the magnetisation in YIG and the polarisation of the spin current generated by the spin Nernst effect in Pt, the latter can or cannot be absorbed at the Pt/YIG interface. This leads, via the absence or presence, respectively, of a spin current backflow from the interface and its conversion into a charge current via the inverse spin Hall effect, to a modulation of the Seebeck signal of the Pt film, the *spin Nernst magnetothermopower* (SMT).

The magnitude of this modulation ΔV is proportional to both, the spin Hall as well as the spin Nernst angle of Pt, given by the ratios $\mathcal{L}_{zx}^{sc,y}/\mathcal{L}_{xx}^{cc} = \theta_{SH}$ and $\mathcal{L}_{zx}^{sq,y}/\mathcal{L}_{xx}^{cq} = \theta_{SN}$ and expressing the efficiency of conversion of a longitudinal charge current into a transverse spin-polarised one, generated by an electric field or a temperature gradient, respectively. Since, using the axis convention of the experiment, the conversion of the y -polarised spin current along z , generated by the SNE, back into an electric field along x is expressed by the inverse spin Hall conductivity $\sigma_{xz}^{i,y} = \sigma_{zx}^y = -\sigma_{zx}^y$ (see Ref. [39]), the relevant (inverse) spin Hall angle θ_{SH} is the ratio $\mathcal{L}_{xz}^{sc,y}/\mathcal{L}_{xx}^{cc} = -\mathcal{L}_{zx}^{sc,y}/\mathcal{L}_{xx}^{cc}$.

Using $\mathbf{E} = \underline{S}\nabla T$ and the expressions for the electrical and thermoelectrical conductivities in terms of Eqs. (8) and (9), σ_{xx} (σ_{zx}^y) = $-e\mathcal{L}_{xx}^{cc}$ ($-e\mathcal{L}_{zx}^{sc,y}$) and α_{xx} (α_{zx}^y) = $-\frac{1}{T}\mathcal{L}_{xx}^{cq}$ ($-\frac{1}{T}\mathcal{L}_{zx}^{sq,y}$), their (temperature-dependent) ratio can be written as

$$\frac{\theta_{SH}}{\theta_{SN}}(T) = \frac{-\sigma_{zx}^y(T) \alpha_{xx}(T)}{\sigma_{xx}(T) \alpha_{zx}^y(T)} = +S_{xx}(T) \frac{\sigma_{zx}^y(T)}{\alpha_{zx}^y(T)}. \quad (16)$$

The energy-dependent conductivities entering Eqs. (8) and (9) and the corresponding expressions for the longitudinal transport coefficients, were calculated for a set of energy points E around E_F for each temperature T accounting for the effect of uncorrelated lattice displacements via the alloy-analogy model [15].

The temperature-dependent spin Hall and Nernst conductivities subsequently obtained from these via Eqs. (8) and (9) are shown in Fig. (5) as circles and triangles, respectively. The ratio of the spin Hall and the spin Nernst angle, expressed by the above quantities as in Eq. (16), is shown as a function of temperature as diamonds (right y axis). As can be seen, for the conventions and definitions chosen here, the two angles have opposite signs for the whole temperature range and $\theta_{SN} > \theta_{SH}$ for $T > 210$ K.

If one extends Eq. (7) and the corresponding expression for the charge current by an additional spin force $\mathbf{F}^s$, exerted on a given system, e.g., due to the presence of a spin accumulation, and the corresponding response coefficients, the Seebeck coefficient for open-circuit conditions (for both, charge and spin) contains additional contributions from the response coefficients

Figure 5. (Color online) Temperature dependence of the spin Hall and the spin Nernst conductivity in bulk fcc-Pt, σ_{zx}^y and α_{zx}^y , respectively, and of the ratio θ_{SH}/θ_{SN} as defined in Eq. (16).

$\mathcal{L}^{sc}$ (and its Onsager reciprocal $\mathcal{L}^{cs}$) and $\mathcal{L}^{sq}$. For a system having the magnetic Laue group $m\bar{3}m1'$ like the Platinum strip considered here, the explicit expression for the relation between an electric field generated in x direction by a temperature gradient $\nabla_x T$ reads

$$\frac{E_x}{\nabla_x T} = S_{xx} = -\frac{1}{eT} \frac{\mathcal{L}_{xx}^{cc}\mathcal{L}_{xx}^{cq} - \mathcal{L}_{xz}^{cs,y}\mathcal{L}_{zx}^{sq,y}}{(\mathcal{L}_{xx}^{cc})^2 + (\mathcal{L}_{zx}^{sc,y})^2}. \quad (17)$$

Obviously, the second term in the numerator describes the spin Nernst magnetothermopower.

Figure 6. (Color online) Difference between the charge Seebeck coefficient for open and closed boundary conditions for charge and spin currents, $\Delta S = S - S^{\text{spin}}$, in bulk fcc-Pt and its relative magnitude compared to S .

Figure 6 shows the temperature dependence of $\Delta S = S - S^{\text{spin}}$ and its relative magnitude compared to the conventional Seebeck coefficient S for closed boundary conditions. It should be noted that interface effects are completely neglected here, nevertheless $\Delta S/S \approx -1.7 \cdot 10^3$ is in good agreement with the relative SMT signal $\Delta V/V = -1.5 \cdot 10^3$ of the experiment [52]. This suggests that the absorption of the spin current at the interface due to the spin mixing conductance or spin transfer torque should be highly efficient.

Figure 7. Family of spin-orbit torque (SOT) related effects as exemplified on Mn/W magnetic bilayer with broken structural inversion symmetry. (a) SOT: An applied in-plane electric field $\mathbf{E}$ generates a torque $\boldsymbol{\tau}$ on the magnetization. $\hat{\mathbf{n}}$ is the magnetization direction. (b) Inverse SOT (ISOT): Magnetization dynamics $\partial \hat{\mathbf{n}} / \partial t$ drives an in-plane electric current $\mathbf{J}$. (c) Thermal SOT (TSOT): The application of a temperature gradient ∇T generates a torque on the magnetization $\boldsymbol{\tau}$. (d) Inverse thermal SOT (ITSOT): Magnetization dynamics drives an in-plane heat current $\mathbf{J}_Q$. Figure taken from [68].

4. Direct and Inverse Thermal Spin-Orbit Torques

While spin-transfer torques rely on the exchange of spin angular momentum between two magnets with different magnetization directions, the so-called spin-orbit torques (SOTs) have been discovered only recently [57, 58, 59] and they are attributed to the spin-orbit-mediated exchange of angular momentum between the crystal lattice and the magnetization, see Fig. 7(a). SOTs exist also in systems with collinear magnetization when inversion symmetry is broken, and it has been shown that SOTs can lead to a reversal of a ferromagnetic magnetization without the help of an additional polarizing layer. [60, 61, 62] Moreover, SOTs were shown to lead to a very fast domain wall motion in thin films at low current density. [63, 58, 64] This suggests that SOTs could play a crucial role in the next generation of spintronics devices. Recently, also the effect of the inverse spin-orbit torque (ISOT) has been uncovered, which consists in the production of an electric current due to magnetization dynamics, see Fig. 7(b) [65, 66, 67].

While the idea of replacing the electric field by a temperature gradient in transport experiments has already inspired research on a plethora of phenomena such as the anomalous Nernst and the spin Nernst effect (SNE), which are at the core of this review

article, the option to use thermal gradients for driving the magnetization dynamics in ferromagnets remains to be explored in-depth. These phenomena, which are thermal analogues of SOT and ISOT, have been suggested theoretically very recently [69, 68]. Namely, within the setup of the thermal SOT (TSOT) the application of a temperature gradient results in a torque on the magnetization, Fig. 7(c) [69], and, conversely, the inverse thermal SOT (ITSOT), Fig. 7(d), is responsible for driving heat currents by magnetization dynamics in ferromagnets [68]. Below, we review the theory of the latter promising thermal effects and present examples of *ab-initio* calculations, which allow us to make realistic estimates of the magnitude of these novel phenomena in magnetic bilayers.

Electrical Spin-Orbit Torques. The expressions for the electric field driven SOTs can be obtained from the Kubo linear response formalism, and they can be evaluated within density functional theory in order to obtain realistic theoretical *ab-initio* predictions of these effects [70]. Within linear response the torque $\boldsymbol{\tau}$ exerted on the ferromagnetic magnetization when an electric field $\mathbf{E}$ is applied is given by $\boldsymbol{\tau} = \mathbf{t}\mathbf{E}$. The torkance tensor $\mathbf{t}$ has three contributions: [70]

$$\begin{aligned} t_{ij}^{(a)} &= -\frac{e}{h} \int_{-\infty}^{\infty} dE \frac{\partial f^0(E)}{\partial E} \text{Tr} \langle \mathcal{T}_i \mathcal{G}^+(E) v_j \mathcal{G}^-(E) \rangle, \\ t_{ij}^{(b)} &= \frac{e}{h} \int_{-\infty}^{\infty} dE \frac{\partial f^0(E)}{\partial E} \Re \text{Tr} \langle \mathcal{T}_i \mathcal{G}^+(E) v_j \mathcal{G}^+(E) \rangle, \\ t_{ij}^{\Pi} &= \frac{e}{h} \int_{-\infty}^{\infty} dE f^0(E) \Re \text{Tr} \langle \mathcal{T}_i \mathcal{G}^+(E) v_j \frac{d\mathcal{G}^+(E)}{dE} \\ &\quad - \mathcal{T}_i \frac{d\mathcal{G}^+(E)}{dE} v_j \mathcal{G}^+(E) \rangle, \end{aligned} \quad (18)$$

with $\mathcal{G}^+(E)$ and $\mathcal{G}^-(E)$ as retarded and advanced Green functions, v_j as the j th cartesian component of the velocity operator, $\mathcal{T}_i$ as the i th cartesian component of the torque operator and $f^0(E)$ as the Fermi distribution function. The torque operator is given by $\mathcal{T}(\mathbf{r}) = \mathbf{m} \times \hat{\mathbf{n}} \Delta^{\text{xc}}(\mathbf{r})$, where $\hat{\mathbf{n}}$ is the direction of magnetization and $\Delta^{\text{xc}}(\mathbf{r}) = \frac{1}{2\mu_B} [V_{\text{minority}}^{\text{eff}}(\mathbf{r}) - V_{\text{majority}}^{\text{eff}}(\mathbf{r})]$ is the exchange field, i.e., the difference between the potentials of minority and majority electrons. $\mathbf{m} = -\mu_B \boldsymbol{\sigma}$ is the spin magnetic moment operator, μ_B is the Bohr magneton and $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)^T$ is the vector of Pauli spin matrices. The first two terms, $t_{ij}^{(a)}$ and $t_{ij}^{(b)}$, are Fermi surface terms, because $\partial f^0 / \partial E$ is nonzero only in a small region around the Fermi surface. The third term, t_{ij}^{Π} , is a Fermi sea term, because all occupied states contribute to it. The brackets $\langle \dots \rangle$ denote again the configurational average as in Eq. (11). In the bilayer geometry considered here, $\mathbf{E}$ is in the plane of the bilayer, while $\boldsymbol{\tau}$ can point in any direction, depending

on the magnetization direction. Therefore, $\mathbf{t}$ is a 3×2 matrix in the case of the bilayers considered here.

The torkance tensor depends generally on the magnetization direction, i.e., $\mathbf{t} = \mathbf{t}(\hat{\mathbf{n}})$, and can be decomposed into even and odd components with respect to magnetization reversal, i.e., $t_{ij}(\hat{\mathbf{n}}) = t_{ij}^{\text{even}}(\hat{\mathbf{n}}) + t_{ij}^{\text{odd}}(\hat{\mathbf{n}})$. For the symmetry of the bilayers discussed in this section (FePt/Pt(001) and Mn/W(001) where we use the convention that the z axis points out-of-plane, while the x and y axes coincide with the [100] and [010] in-plane directions), see Fig. 7, it is easy to show that $t_{xx}(\hat{\mathbf{e}}_z) = t_{yy}(\hat{\mathbf{e}}_z) = t_{xx}^{\text{odd}}(\hat{\mathbf{e}}_z)$ and $t_{yx}(\hat{\mathbf{e}}_z) = -t_{xy}(\hat{\mathbf{e}}_z) = t_{yx}^{\text{even}}(\hat{\mathbf{e}}_z)$, when the magnetization points in z -direction, which we denote by the unit-vector $\hat{\mathbf{e}}_z$. Consequently, $t_{xx}^{\text{even}}(\hat{\mathbf{e}}_z) = 0$ and $t_{yx}^{\text{odd}}(\hat{\mathbf{e}}_z) = 0$. Ignoring higher-order anisotropies of SOT, we can express $\boldsymbol{\tau}$ for general magnetization direction $\hat{\mathbf{n}}$ as

$$\boldsymbol{\tau} = t_{xx}^{\text{odd}}(\hat{\mathbf{e}}_z) \hat{\mathbf{n}} \times (\mathbf{E} \times \hat{\mathbf{e}}_z) + t_{yx}^{\text{even}}(\hat{\mathbf{e}}_z) \hat{\mathbf{n}} \times [\hat{\mathbf{n}} \times (\mathbf{E} \times \hat{\mathbf{e}}_z)]. \quad (19)$$

While more sophisticated schemes for treating the effect of disorder on the SOT have been used [71, 72, 73], it appears that in many cases it is sufficient to model the influence of disorder in the system by a constant effective band broadening. Within this model the retarded and advanced Green functions are given by $G^{\text{R}}(E) = \hbar[E - H + i\Sigma]^{-1}$ and $G^{\text{A}}(E) = \hbar[E - H - i\Sigma]^{-1}$, with parameter Σ characterizing the disorder strength. In the clean limit of vanishing disorder $\Sigma \rightarrow 0$ the even and odd components of the torkance tensor acquire qualitatively different forms:

$$t_{ij}^{\text{even}} = -\frac{2e}{\mathcal{N}} \hat{\mathbf{e}}_i \cdot \sum_{\mathbf{k}, n} f^0(\epsilon_{\mathbf{k}n}) \left[\hat{\mathbf{n}} \times \Im \left\langle \frac{\partial u_{\mathbf{k}n}}{\partial \hat{\mathbf{n}}} \middle| \frac{\partial u_{\mathbf{k}n}}{\partial k_j} \right\rangle \right], \quad (20)$$

and

$$t_{ij}^{\text{odd}} = \frac{e\hbar}{2\Sigma\mathcal{N}} \sum_{\mathbf{k}n} \langle \psi_{\mathbf{k}n} | \mathcal{T}_i | \psi_{\mathbf{k}n} \rangle \langle \psi_{\mathbf{k}n} | v_j | \psi_{\mathbf{k}n} \rangle \frac{\partial f^0(\epsilon_{\mathbf{k}n})}{\partial \epsilon_{\mathbf{k}n}}, \quad (21)$$

where the $\mathbf{k}$ -vector $\mathbf{k}$ is a Bloch vector in the Brillouin zone, $\mathcal{N}$ is the number of $\mathbf{k}$ -points, n runs over all bands, $\epsilon_{\mathbf{k}n}$ is the band energy, $\psi_{\mathbf{k}n}$ and $u_{\mathbf{k}n}$ are the Bloch states and their lattice-periodic parts, respectively, and $\hat{\mathbf{e}}_i$ is the unit vector along the i th cartesian direction. In this limit the even torkance has the form of a Berry curvature and it is independent of Σ in the limit of $\Sigma \rightarrow 0$. It constitutes the intrinsic contribution to the torkance, and it is analogous to the intrinsic anomalous or spin Hall effects. The odd part of the torkance, on the other hand, diverges like $1/\Sigma$ in metals in the limit of small Σ , i.e., it is proportional to the quasi-particle lifetime similar to the diagonal electrical conductivity [74], and it is thus strongly

dependent on the amount of scattering present in the system. One contribution to the odd torque arises from the Edelstein effect [75]. In the remainder of this subsection we discuss the calculations at $\Sigma = 25$ meV, which corresponds to experiments performed at room temperature, if the main source of disorder in the system is e.g. due to phonons. The electronic structure was calculated within the Generalized Gradient Approximation to DFT using the Full-Potential Linearized Augmented Plane Wave method as implemented in the code FLEUR [76]. For the SOT calculations, the WANNIER90 code was employed [77, 78] in order to compute maximally localized Wannier functions for a numerically inexpensive but accurate interpolation of the band structure.

Thermal Spin-Orbit Torques. Similarly to the spin Hall or anomalous Hall conductivities, which are used to quantify the Hall effects, the torkance describes the SOT arising from an applied electric field, i.e., it corresponds to the situation in which the torque is driven by a mechanical force. A torque can also be induced by a temperature gradient ∇T , i.e., it can also originate from statistical forces. Within linear response this thermal torque reads:

$$\boldsymbol{\tau} = -\beta \nabla T, \quad (22)$$

where β is the so-called *thermal torkance*. In analogy to the torkance driven by electrical currents, the thermal torkance can be decomposed into even and odd components with respect to the magnetization direction, and it has the same symmetry properties. The intrinsic even part of the thermal torkance is analogous to the intrinsic anomalous Nernst [79, 80] and spin Nernst conductivities [51, 81, 53, 54]. Similar to the latter effects, it can be shown that the thermal torkance β can be computed directly from its mechanical counterpart employing a Mott-like relation: [82]

$$\beta_{ij}(T) = -\frac{1}{e} \int dE \frac{\partial f^0(E, \mu, T)}{\partial \mu} t_{ij}(E) \frac{E - \mu}{T}, \quad (23)$$

where $t_{ij}(E)$ is the torkance tensor with Fermi energy set to E and μ is the chemical potential. For the bilayers discussed here, β_{ij} is a 3×2 matrix, because we consider only in-plane temperature gradients.

An example of accessing the TSOT from first principles according to Eq. 23 has been given in Ref. [69]. In the latter work, Géranton and co-authors consider 2 layers of L1₀-FePt oriented along [001]-axis and terminated with Fe atoms (Fe/Pt/Fe/Pt/Fe) deposited on the upper side of a Pt(001) film, where thicknesses of 6, 12 and 18 Pt layers were considered. The TSOT in this system was computed according to Eq. (23) at the temperature of $T = 300$ K using as input the calculated energy

Figure 8. a) Even thermal torkance β_{yx}^{even} and b) odd thermal torkance β_{xx}^{odd} are calculated for $T = 300$ K for $\text{L1}_0\text{-FePt/Pt}$ thin films with $N=6, 12, 18$ Pt layers using Eq. (23) in Ref. [[69]]. The line of circles in the upper figure corresponds to the even thermal torkance β_{yx}^{SNE} estimated from the spin Nernst conductivity of bulk fcc Pt, Eq. (25).

dependence of the even and odd torkances computed at $\Sigma = 25$ meV. The computed energy dependence of the even and odd thermal torkances β_{yx}^{even} and β_{xx}^{odd} of $\text{L1}_0\text{-FePt/Pt}$ thin films is shown in Fig. 8. The largest values of β_{yx}^{even} of the order of tens of $\mu\text{eV}\cdot\text{a}_0\cdot\text{K}^{-1}$ are achieved around the Fermi energy, while the magnitude of β_{xx}^{odd} is maximal away from the Fermi energy, reaching as much as $100\mu\text{eV}\cdot\text{a}_0\cdot\text{K}^{-1}$. The thermal torkances for 6 and 12/18 layers of Pt substrate differ in sign in a region around -0.4eV and the difference between thermal torkances for 12 and 18 layers becomes more pronounced. At the true Fermi energy, β_{xx}^{odd} exhibits a change of sign when changing the Pt thickness. This emphasizes the extreme sensitivity of the thermal torkance to the parameters which determine the electronic structure of the system.

It is known that in the systems of the bilayer type depicted in Fig. 7, the dominant contribution to the even torkance is often provided by the spin Hall current which is generated in the substrate upon the application of the electric field [70]. Within the setup of the spin Nernst effect (SNE) [51, 81], whose properties can be characterized by the spin Nernst conductivity (SNC) α , an applied temperature gradient will result in transverse spin current. Keeping in mind the geometry

of bilayers, the relationship between a temperature gradient applied along the x axis and the spin current density with spin-polarization along the y axis which propagates along the z axis, reads:

$$j_z^y = -\alpha_{zx}^y \nabla_x T, \quad (24)$$

where the the SNC is given by the Mott-type relation

$$\alpha_{zx}^y = -\frac{1}{e} \int dE \frac{\partial f^0(E, \mu, T)}{\partial \mu} \sigma_{zx}^y(E) \frac{E - \mu}{T}, \quad (25)$$

and $\sigma_{zx}^y(E)$ is the corresponding energy-dependent component of the spin Hall conductivity. We define the spin Nernst thermal torkance β_{yx}^{SNE} as the torkance that arises hypothetically from the absorption of the entire generated spin Nernst current by the magnetization in the magnetic overlayer. This spin Nernst thermal torkance of FePt/Pt is shown in Fig. 8 as a function of the position of the Fermi energy together with β_{yx}^{even} . By comparing the two torkances we can conclude that in this system the overall behavior of β_{yx}^{even} with energy is in accordance with that of β_{yx}^{SNE} in a wide window of energies. This underlines a close correlation between the phenomena of TSOT and the spin Nernst effect.

In order to get a feeling for the magnitude of the intrinsic thermal torque that can be achieved realistically in FePt/Pt bilayers, we determine the temperature gradient $|\nabla T|^0$ that is required to achieve the same effective magnetic field as is produced by the electrical SOT with a current density of $j \sim 10^7 \text{ A/cm}^2$, which is typically needed for magnetization-switching by SOT in similar magnetic bilayer systems. The value of $|\nabla T|^0$ of the order of 2 K/nm which can be obtained for $\text{L1}_0\text{-FePt/Pt}$ bilayers at their true Fermi energy is one order of magnitude larger than what can be achieved experimentally in this type of systems. [83] Therefore, we may expect that the intrinsic thermal SOT is experimentally observable in FePt/Pt bilayers, but that it is probably too small to be used for switching the magnetization.

Moreover, one can conclude that at the current level of experimental techniques the TSOT can be made as large as the electrical SOT by proper electronic structure engineering, which can go along various paths. Firstly, as apparent from Fig. 8, for FePt/Pt bilayers the thermal torkances can be an order of magnitude larger if the Fermi energy is set to $\sim 0.6 \text{ eV}$ above its true value - this corresponds roughly to using e.g. $\text{L1}_0\text{-(Fe}_{1-x}\text{Co}_x\text{)Pt/Pt}_{1-x}\text{Au}_x$ instead of FePt/Pt , with $x \sim 0.6$ if a constant density of states of $\sim 1 \text{ eV}^{-1}$ per atom for $\text{Fe}_{1-x}\text{Co}_x\text{Pt}$ and $\text{Pt}_{1-x}\text{Au}_x$ is assumed.[69] Secondly, the calculations presented in Ref. [[69]] show that upon decreasing the disorder strength Σ the energy dependence of the odd and, particularly, even torkances exhibits strong deviations

Figure 9. Intrinsic spin Nernst conductivity in selected non-magnetic bulk metals as computed from *ab-initio* according to Eq. 25 at T=291K.

from the smooth behavior shown above, acquiring sharp features and sign changes at the scale of tens of meVs. This effect is due to the fine features in the electronic structure of thin films, which get promoted as the band broadening is decreased. Correspondingly, upon reducing the degree of disorder in the system (e.g. by lowering of the temperature or concentration of impurities) the magnitude of the TSOT, qualitatively proportional to the degree of raggedness of the torkance as a function of energy, can be significantly enhanced. Since the SNE is directly proportional to the degree of the changes that the SHC experiences around the Fermi energy, a promising way of engineering the magnitude of the SNE is to exploit the drastic effect that the impurity scattering in the dilute limit can have on the energy-dependence of the spin Hall effect. [84] Overall, tuning the details of disorder by alloying, phonons, magnetic excitations, substitutional disorder etc. can provide a fruitful path towards technologically relevant applications of the thermal spin-orbit torque.

Finally, the close correlation between the TSOT and the SNE that we outlined is a compelling force to consider substrates with a larger spin Nernst conductivity than that of fcc Pt as another opportunity to maximize the TSOT. Fig. 9 shows the intrinsic spin Nernst conductivity of selected simple non-magnetic metals as computed from Eq. 25. Clearly, in particular fcc Pd and Rh stick out as promising substrates for sizeable TSOT, since the intrinsic SNCs for these metals constitute +20804 (Pd), and -20779 ($\hbar/e$) $\mu\text{A}\cdot\text{cm}^{-1}\cdot\text{K}^{-1}$ (Rh), which is larger in magnitude than the SNC of fcc Pt of -8383 ($\hbar/e$) $\mu\text{A}\cdot\text{cm}^{-1}\cdot\text{K}^{-1}$ by a factor of 2.5. [69] Recently, the SNE has been observed experimentally in Pt by measuring the spin Nernst magneto-thermopower

(SMT). [85] Therefore, the large theoretical values of the intrinsic SNE in Pd and Rh are relevant not only to the TSOT, but also to the SMT.

Inverse thermal spin-orbit torque (ITSOT). The effect of inverse thermal spin-orbit torque consists in the generation of heat current by magnetization dynamics in ferromagnets with broken inversion symmetry and SOI. This effect is reciprocal to the TSOT effect discussed above. Generally, the effect of magnetization dynamics can be described by the time-dependent perturbation δH to the Hamiltonian H [70]

$$\delta H = \frac{\sin(\omega t)}{\omega} \left[\hat{\mathbf{n}} \times \frac{\partial \hat{\mathbf{n}}}{\partial t} \right] \cdot \boldsymbol{\tau}, \quad (26)$$

and correspondingly, the effect of ITSOT is encompassed by the following expression for the heat current $\mathcal{Q}$ driven by the magnetization dynamics:

$$\mathcal{Q} = -\tilde{\beta} \left[\hat{\mathbf{n}} \times \frac{\partial \hat{\mathbf{n}}}{\partial t} \right], \quad (27)$$

where the total $\tilde{\beta}_{ij}(\hat{\mathbf{n}})$ coefficient is related to the total torkance $t_{ji}(-\hat{\mathbf{n}}, E)$ for magnetization in $-\hat{\mathbf{n}}$ direction by

$$\tilde{\beta}_{ij}(\hat{\mathbf{n}}) = -\frac{1}{eV} \int_{-\infty}^{\infty} dE \frac{\partial f(E, \mu, T)}{\partial \mu} (E - \mu) t_{ji}(-\hat{\mathbf{n}}, E). \quad (28)$$

Clearly, the ITSOT described by (28) is the Onsager-reciprocal of the TSOT, Eq. (23). One of the consequences of this is that a part of the even contribution to the driven heat current is due to the inverse spin Nernst effect, which gives rise to a transverse flow of heat in response to the propagation of a spin current. Direct comparison of Eq. (28) and Eq. (23) yields

$$\beta(\hat{\mathbf{n}}) = -\frac{V}{T} [\tilde{\beta}(-\hat{\mathbf{n}})]^T \quad (29)$$

and thus one can write a general relation:

$$\begin{pmatrix} -\mathcal{Q} \\ \tau/V \end{pmatrix} = \begin{pmatrix} T\lambda(\hat{\mathbf{n}}) & \tilde{\beta}(\hat{\mathbf{n}}) \\ [\tilde{\beta}(-\hat{\mathbf{n}})]^T & -\Lambda(\hat{\mathbf{n}}) \end{pmatrix} \begin{pmatrix} \frac{\nabla T}{T} \\ \hat{\mathbf{n}} \times \frac{\partial \hat{\mathbf{n}}}{\partial t} \end{pmatrix}, \quad (30)$$

where λ is the thermal conductivity tensor and Λ describes Gilbert damping and gyromagnetic ratio [65]. As expected, the response matrix

$$\mathcal{A}(\hat{\mathbf{n}}) = \begin{pmatrix} T\lambda(\hat{\mathbf{n}}) & \tilde{\beta}(\hat{\mathbf{n}}) \\ [\tilde{\beta}(-\hat{\mathbf{n}})]^T & -\Lambda(\hat{\mathbf{n}}) \end{pmatrix} \quad (31)$$

satisfies the Onsager symmetry $\mathcal{A}(\hat{\mathbf{n}}) = [\mathcal{A}(-\hat{\mathbf{n}})]^T$.

The TSOT and ITSOT were investigated from *ab-initio* in a Mn/W(001) magnetic bilayer composed of

Figure 10. Thermal torkance β vs. temperature of a Mn/W(001) magnetic bilayer for magnetization in z direction. Solid line: Even component β_{yx}^{even} of the thermal torkance. Dashed line: Odd component β_{xx}^{odd} of the thermal torkance. β is plotted in units of $\mu\text{eV}a_0/\text{K} = 8.478 \times 10^{-36} \text{Jm/K}$, where a_0 is Bohr's radius.

one monolayer of Mn deposited on 9 layers of W(001) in Ref. [[68]], while the SOT in this system has been already discussed by Freimuth et al. [70] In order to obtain TSOT and ITSOT, the torkance was computed first in order to determine the TSOT and ITSOT coefficients β and $\tilde{\beta}$, respectively. In analogy to the case considered above, due to symmetry it is sufficient to discuss the TSOT coefficients β_{yx}^{even} and β_{xx}^{odd} , which are shown in Fig. 10 as a function of temperature. For small temperatures we find $\beta_{ij} \propto T$ as expected from

$$\beta_{ij} \simeq -\frac{\pi^2 k_B^2 T}{3e} \frac{\partial t_{ij}}{\partial \mu}, \quad (32)$$

which is obtained from (23) using the Sommerfeld expansion. Slightly above 100K both β_{yx}^{even} and β_{xx}^{odd} stop following the linear behavior of the low temperature expansion (32): After reaching a maximum both β_{yx}^{even} and β_{xx}^{odd} decrease and finally change sign. At $T = 300\text{K}$ the thermal torkances are $\beta_{yx}^{\text{even}} = 5.24 \times 10^{-35} \text{Jm/K}$ and $\beta_{xx}^{\text{odd}} = -3.21 \times 10^{-36} \text{Jm/K}$. These thermal torkances are of comparable magnitude to those shown for FePt/Pt bilayer above.[69] Correspondingly, the values of $\tilde{\beta}_{yx}^{\text{even}} = -99.49 \mu\text{J/m}^2$ and $\tilde{\beta}_{xx}^{\text{odd}} = -6.09 \mu\text{J/m}^2$ at $T = 300\text{K}$ are obtained.

When the magnetization precesses around the z axis in ferromagnetic resonance (this situation is sketched in Figure 1d) with frequency ω and cone angle θ according to

$$\hat{n}(t) = [\sin(\theta) \cos(\omega t), \sin(\theta) \sin(\omega t), \cos(\theta)]^T, \quad (33)$$

the following expressions for the ITSOT heat current

can be obtained from (27) in the limit of small θ :

$$\begin{aligned} (\mathcal{Q})_x &= \omega \theta \left[\tilde{\beta}_{xx}^{\text{odd}} \cos(\omega t) - \tilde{\beta}_{yx}^{\text{even}} \sin(\omega t) \right] \\ (\mathcal{Q})_y &= \omega \theta \left[\tilde{\beta}_{yx}^{\text{even}} \cos(\omega t) + \tilde{\beta}_{xx}^{\text{odd}} \sin(\omega t) \right], \end{aligned} \quad (34)$$

where we used $\tilde{\beta}_{xx} = \tilde{\beta}_{yy} = \tilde{\beta}_{xx}^{\text{odd}}$ and $-\tilde{\beta}_{xy} = \tilde{\beta}_{yx} = \tilde{\beta}_{yx}^{\text{even}}$ obtained from symmetry considerations. Using the ITSOT coefficients $\tilde{\beta}_{yx}^{\text{even}}$ and $\tilde{\beta}_{xx}^{\text{odd}}$ one can determine the amplitudes of $(\mathcal{Q})_x$ and $(\mathcal{Q})_y$. Assuming a cone angle of 1° and a frequency of $\omega = 2\pi \cdot 5\text{GHz}$ the amplitude of the oscillating heat current density $(\mathcal{Q})_x$ can be estimated to be $\approx 55 \frac{\text{kW}}{\text{m}^2}$. The heat current density $(\mathcal{Q})_y$ has the same amplitude. Using the thermal conductivity of bulk W of $\lambda_{xx} = 174 \text{ W/(Km)}$ [86] at $T = 300 \text{ K}$ one arrives at an estimate of the temperature gradient needed to drive a heat current of this magnitude of 316 K/m . Taking into account the finite thickness of the Mn/W(001) film the amplitude of the heat current per length in x direction is thus of the order of $100 \mu\text{W/m}$. This suggests that $\mathcal{Q}$ should be detectable in ferromagnetic resonance experiments on Mn/W bilayers.

According to (34) the heat current can be made larger by increasing the cone angle. However, in ferromagnetic resonance experiments the cone angle θ is small. Therefore, exploring an alternative idea, Freimuth and co-workers estimate the heat current driven by a flat cycloidal spin spiral with the spin-spiral vector q that moves with velocity w in x direction, arriving at an estimate for $(\mathcal{Q})_x = -\tilde{\beta}_{xy}^{\text{even}} \omega q$ as obtained from (27), i.e., a constant-in-time heat current in x direction. Using the value for $\tilde{\beta}_{xy}^{\text{even}}$ from above and assuming a spin-spiral wavelength of 2.3nm [87] one obtains a heat current density of $(\mathcal{Q})_x = -270 \text{ kW/m}^2$ for a spin spiral moving with a speed of $w = 1 \text{ ms}^{-1}$. This estimate suggests that fast domain walls moving at a speed of the order of 100 ms^{-1} can be used to drive significant heat currents via the effect of ITSOT, that correspond to temperature gradients of the order of $0.1 \text{ K}/(\mu\text{m})$.

5. Semiclassical approach for spin-orbit driven thermal transport

5.1. Introduction

Recently, the field of *spin caloritronics* [2, 3] emerged, which couples the known technologies thermoelectrics and spintronics, leading to a promising class of new effects. Thermoelectric phenomena, combining the electrical and thermal aspects of charge transport, have been studied for many years. Among them, the Seebeck effect and the Nernst effect were observed and well understood. In the last years, the field of spintronics gained importance in the course of storage

technology development. [88] The aims are the control of the electron's spin degree of freedom and the creation of spin currents either by an electric field or via spin pumping. Now, spin caloritronics handles phenomena, where the spin degree of freedom is influenced by thermal gradients. As a first prominent phenomenon of this type, the spin Seebeck effect was observed. [89, 90, 91] There a temperature gradient was applied to a ferromagnetic strip while a linearly varying transverse voltage in Pt contacts was measured via the inverse spin Hall effect. [92] Since the effect was not only observed in metals, but later in magnetic insulators and semiconductors, [5, 93] it became finally clear that the phenomenon is caused by magnons. [94, 95] The main focus of this article is related to the spin Nernst effect (SNE) [96, 81, 97, 53, 98, 99, 100, 54, 84, 85], which is the thermal analog to the intensively studied spin Hall effect (SHE) [101, 102, 103]. The fingerprint of both phenomena is a transverse spin current or spin accumulation. However, whereas the SHE is caused by an external electric field, the SNE occurs due to an applied temperature gradient. For its description we employ here the concept of spin-dependent transport coefficients, [90] which was successfully used to describe the thermally driven spin injection from a ferromagnet to a nonmagnetic material. [83]

In our recent paper, [53] an *ab-initio* study of the extrinsic SNE in Cu-based dilute alloys was presented. Here, we perform a detailed consideration of the theoretical basis for the discussed phenomenon, turning the attention to its microscopic origins. We provide a minute description of the used approach, which is based on the semiclassical Boltzmann theory, and give insight into the mechanisms influencing the obtained results.

5.2. Transport equation

The derivation of the transport equation provided by this section is mostly reproducing the corresponding approach of Ref. [1]. However, in comparison to this reference we go beyond the anisotropic relaxation time approximation for the mean free path. In addition, some features of the relativistic treatment, caused by the presence of spin-orbit coupling, require a generalized consideration of the scattering term in the Boltzmann equation. For simplicity, in this Section the spin degree of freedom for an electron is combined with the band index. However, it will be used explicitly in Section III for the two-current model.

The extrinsic skew-scattering mechanism [104, 105, 50, 106] provides the dominating contribution to the transverse spin current for dilute alloys [107, 38, 108, 109]. This mechanism, reflecting an asymmetric scattering of electrons with opposite spin orientations caused by spin-orbit coupling, can be well described

within the semiclassical theory. [38] It means we use the Boltzmann equation for the distribution function $f_k(\mathbf{r})$

$$\frac{\partial f_k}{\partial t} + \dot{\mathbf{r}}_k \frac{\partial f_k}{\partial \mathbf{r}} + \dot{\mathbf{k}} \frac{\partial f_k}{\partial \mathbf{k}} = \left(\frac{\partial f_k}{\partial t} \right)_{\text{scatt.}}, \quad (35)$$

where $\dot{\mathbf{r}}_k \equiv \mathbf{v}_k = (1/\hbar)(\partial E_k/\partial \mathbf{k})$ is the group velocity. Here k is a shorthand notation for the crystal momentum $\mathbf{k}$ and the band index n . Further on, the first and third terms on the left hand side of Eq. (35) can be skipped, since we are interested in steady-state solutions without external fields. Instead, we take into account an applied temperature gradient ∇T . Moreover, we restrict our consideration to homogeneous systems. It implies that only the nonequilibrium part g_k of the distribution function $f_k(\mathbf{r}) = f_k^0 + g_k(\mathbf{r})$ shows a spatial dependence. The corresponding equilibrium part is given by the Fermi-Dirac function

$$f_k^0 = \{\exp[(E_k - \mu)/k_B T] + 1\}^{-1}. \quad (36)$$

Following the arguments of Ziman, [1] one can write the nonequilibrium distribution function as

$$f_k(\mathbf{r}) = f_k^0(T(\mathbf{r}) - \mathbf{\Lambda}_k \cdot \nabla T, \mu(\mathbf{r}) - \mathbf{\Lambda}_k \cdot \nabla \mu), \quad (37)$$

where $\mathbf{\Lambda}_k$ is the vector mean free path. Here, the term with $\nabla \mu$ arises due to the applied temperature gradient which changes the chemical potential μ spatially.

Now we restrict our consideration to the linear response of the system to the thermal perturbation. This means taking into account only the linear terms in the Taylor expansion

$$f_k \approx f_k^0 + \left. \frac{\partial f_k}{\partial (\nabla T)} \right|_{\nabla T=0, \nabla \mu=0} \cdot \nabla T + \left. \frac{\partial f_k}{\partial (\nabla \mu)} \right|_{\nabla T=0, \nabla \mu=0} \cdot \nabla \mu \quad (38)$$

that provides us the nonequilibrium part of the distribution function as

$$g_k = \frac{\partial f_k^0}{\partial E_k} \left[\frac{E_k - \mu}{T} \mathbf{\Lambda}_k \cdot \nabla T + \mathbf{\Lambda}_k \cdot \nabla \mu \right]. \quad (39)$$

This function is required for the scattering term of the Boltzmann equation [110, 111]

$$\left(\frac{\partial f_k}{\partial t} \right)_{\text{scatt.}} = \sum_{k'} (P_{k'k} g_{k'} - P_{kk'} g_k), \quad (40)$$

where the microscopic transition probability

$$P_{kk'} = \frac{2\pi}{\hbar} c_i N |T_{kk'}|^2 \delta(E_k - E_{k'}) \quad (41)$$

for the scattering from an initial state k to a final state k' scales with the impurity concentration c_i and the

total number of atoms N in the system. [14] Here, the transition matrix elements $T_{kk'} \equiv T_{k \rightarrow k'}$ can be obtained from the solution of the impurity problem on a cluster in the real space. [14, 16] The second part in the scattering term can be rewritten using the relaxation time τ_k , the mean time a particle in the state k can move until it is scattered, as

$$\frac{1}{\tau_k} = \sum_{k'} P_{kk'} . \quad (42)$$

Now we come back to the left hand side of Eq. (35), which is represented for the considered phenomenon by

$$\begin{aligned} \dot{\mathbf{r}}_k \frac{\partial f_k}{\partial \mathbf{r}} &= \mathbf{v}_k \left(\frac{\partial f_k^0}{\partial T} \nabla T + \frac{\partial f_k^0}{\partial \mu} \nabla \mu \right) = \\ &= \mathbf{v}_k \frac{\partial f_k^0}{\partial E_k} \left[-\frac{E_k - \mu}{T} \nabla T - \nabla \mu \right] , \end{aligned} \quad (43)$$

neglecting terms of higher order in ∇T and $\nabla \mu$. Taking into account Eq. (39), one obtains the following expression for the mean free path

$$\Lambda_k = \tau_k \left[\mathbf{v}_k + \sum_{k'} P_{k'k} \Lambda_{k'} \right] , \quad (44)$$

which is exactly the same as the corresponding one for the case of an applied electric field. [14, 16] This is the physical background of the Wiedemann-Franz law [112] at the microscopic level. [1] In practice, Eq. (44) can be solved iteratively with $\Lambda_k^0 = \tau_k \mathbf{v}_k$ used normally as a starting value. [14, 113]

Here, we would like to emphasize a detail which is important for any practical calculations. Namely, due to the fact that the SHE and SNE are driven by spin-orbit coupling, the non-relativistic microscopic reversibility $P_{kk'} = P_{k'k}$ is not valid anymore. [16, 114] Therefore, it is crucial to keep the correct form of the scattering-in term given by the last part of Eq. (44) arising from the term $P_{k'k} g_{k'}$ of Eq. (40).

For an applied temperature gradient, the electron current density is given by ($e = |e|$)

$$\mathcal{Q} = -\frac{e}{V} \sum_k \mathbf{v}_k f_k = -\frac{e}{V} \sum_k \mathbf{v}_k g_k \quad (45)$$

with g_k defined by Eq. (39) leading to

$$\mathcal{J} = -\frac{e}{V} \sum_k \frac{\partial f_k^0}{\partial E_k} (\mathbf{v}_k \circ \Lambda_k) \left[\nabla \mu + \frac{E_k - \mu}{T} \nabla T \right] . \quad (46)$$

The transport equation can be rewritten as [1]

$$\mathcal{J} = -\hat{L}_0 \nabla \mu - \frac{1}{T} \hat{L}_1 \nabla T . \quad (47)$$

Similar to the charge current, the heat current density [1]

$$\mathcal{Q} = \frac{1}{V} \sum_k \mathbf{v}_k g_k (E_k - \mu) = \frac{1}{e} \hat{L}_1 \nabla \mu + \frac{1}{eT} \hat{L}_2 \nabla T \quad (48)$$

can be introduced. Finally, the linear transport coefficients in Eqs. (47) and (48) are given by

$$\hat{L}_m = -\frac{1}{e} \int dE \hat{\sigma}(E) \left[-\frac{\partial f^0(E, T)}{\partial E} \right] (E - \mu)^m \quad (49)$$

with $m \in \{0, 1, 2\}$. In the limit $T \rightarrow 0$ we reach the conventional limit $\hat{L}_0 \rightarrow -\hat{\sigma}(\mu)/e$. Here, the energy-dependent electrical conductivity

$$\hat{\sigma}(E) = \frac{e^2}{\hbar(2\pi)^3} \sum_n \iint_{E_k=E} \frac{dS_k}{|\mathbf{v}_k|} \mathbf{v}_k \circ \Lambda_k , \quad (k = \{\mathbf{k}, n\}) \quad (50)$$

is calculated via a sum over the electron bands and an integration over the corresponding isoenergetic surfaces E in $\mathbf{k}$ -space. To obtain Eqs. (47)–(50), the relation

$$\sum_k \rightarrow \frac{V}{(2\pi)^3} \int d\mathbf{k}_\perp \iint_{E_k=E} dS_k = \frac{V}{\hbar(2\pi)^3} \int dE \iint_{E_k=E} \frac{dS_k}{|\mathbf{v}_k|} \quad (51)$$

was used. As mentioned in Ref. [53], for the integral in Eq. (49) only a small area around the Fermi energy $\mathcal{E}_F$ needs to be considered, since the energy derivative of $f^0(E, T)$ has an exponential decay.

5.3. Two current description

We are going to discuss two possible scenarios for an experimental situation. For the first one, we assume that a system under consideration is electrically insulated, which means there are no macroscopic charge or spin currents in the steady state. Instead, accumulations of charges or spins at the edges of the sample can occur. For the second scenario, we allow currents transverse to the applied temperature gradient, which we will always assume to be in the x direction: $\nabla T = (\nabla_x T, 0, 0)$.

For the description of both situations, we use the two current model where the charge current and the spin current are given by

$$\mathcal{J} = \mathcal{J}^+ + \mathcal{J}^- \quad \text{and} \quad \mathcal{J}^s = \mathcal{J}^+ - \mathcal{J}^- , \quad (52)$$

respectively. Here, “+” and “−” indices denote the two relativistic spin channels. [115, 116] Due to the presence of both time and space inversion symmetry in the investigated systems, the transport equation (47) can be considered for each spin channel separately

$$\mathcal{J}^\pm = -\hat{L}_0^\pm \nabla \mu^\pm - \frac{1}{T} \hat{L}_1^\pm \nabla T . \quad (53)$$

By analogy, we can introduce the heat current and the spin heat current

$$\mathbf{Q} = \mathbf{Q}^+ + \mathbf{Q}^- \quad \text{and} \quad \mathbf{Q}^s = \mathbf{Q}^+ - \mathbf{Q}^-, \quad (54)$$

respectively. Again the transport equation (48) can be written for each spin channel separately

$$\mathbf{Q}^\pm = \frac{1}{e} \hat{L}_1^\pm \nabla \mu^\pm + \frac{1}{eT} \hat{L}_2^\pm \nabla T. \quad (55)$$

The linear transport coefficients $\hat{L}_m^\pm$ are calculated using Eq. (49) with

$$\hat{\sigma}^\pm(E) = \frac{e^2}{\hbar(2\pi)^3} \iint_{\mathcal{E}_k=\mathcal{E}} \frac{dS_{\mathbf{k}}}{|\mathbf{v}_{\mathbf{k}}|} \mathbf{v}_{\mathbf{k}} \circ \mathbf{\Lambda}_{\mathbf{k}}^\pm \quad (56)$$

as spin- and energy-dependent conductivity tensor.

For a host system with a cubic symmetry, the spin-dependent tensors have the following structure

$$\hat{\sigma}^\pm = \begin{pmatrix} \sigma_{xx}^\pm & -\sigma_{yx}^\pm & 0 \\ \sigma_{yx}^\pm & \sigma_{xx}^\pm & 0 \\ 0 & 0 & \sigma_{zz}^\pm \end{pmatrix}, \quad \hat{L}^\pm = \begin{pmatrix} L_{xx}^\pm & -L_{yx}^\pm & 0 \\ L_{yx}^\pm & L_{xx}^\pm & 0 \\ 0 & 0 & L_{zz}^\pm \end{pmatrix}, \quad (57)$$

assuming that the spin quantization axis is chosen along the z direction. Additionally, taken into account that for nonmagnetic systems the time reversal symmetry is present, we have

$$\sigma_{xx}^+ = \sigma_{xx}^-, \quad \sigma_{yx}^+ = -\sigma_{yx}^-, \quad \sigma_{zz}^+ = \sigma_{zz}^-; \quad (58)$$

$$L_{xx}^+ = L_{xx}^-, \quad L_{yx}^+ = -L_{yx}^-, \quad L_{zz}^+ = L_{zz}^-.$$

Furthermore, for the considered Cu-based alloys, only one spin-degenerate band occurs at and near the Fermi level. Therefore, the sum over bands in Eq. (44) reduces to the sum over the “+” and “−” spin subbands. Consequently, the spin-dependent mean free path is given by

$$\mathbf{\Lambda}_{\mathbf{k}}^\pm = \tau_{\mathbf{k}}^\pm \left[\mathbf{v}_{\mathbf{k}} + \sum_{\mathbf{k}'} P_{\mathbf{k}'\mathbf{k}}^{\pm\pm} \mathbf{\Lambda}_{\mathbf{k}'}^+ + \sum_{\mathbf{k}'} P_{\mathbf{k}'\mathbf{k}}^{\mp\pm} \mathbf{\Lambda}_{\mathbf{k}'}^- \right], \quad (59)$$

where we have taken into account the relation $\mathbf{v}_{\mathbf{k}}^+ = \mathbf{v}_{\mathbf{k}}^- = \mathbf{v}_{\mathbf{k}}$. The second and third terms on the right hand side of the equation above describe spin-conserving and spin-flipping transitions, respectively. The same separation holds for the relaxation time

$$\frac{1}{\tau_{\mathbf{k}}^\pm} = \sum_{\mathbf{k}'} P_{\mathbf{k}\mathbf{k}'}^{\pm\pm} + \sum_{\mathbf{k}'} P_{\mathbf{k}\mathbf{k}'}^{\mp\pm}. \quad (60)$$

In Ref. [53] the spin-flip processes were neglected. Here, we present results obtained including both spin-conserving and spin-flipping transitions.

5.3.1. Spin accumulation Let us start with the situation where the macroscopic charge current vanishes, because the system is electrically insulated, and assume that the spin current also vanishes ($\mathcal{J} = \mathcal{J}^s = 0$). According to Eq. (52), this requires $\mathcal{J}^+$ and $\mathcal{J}^-$ to vanish as well. However, a heat current is flowing. This can be understood as an exchange of “hot” and “cold” electrons, [1] which are of equal amount but have different properties since they come from two sides of the sample being at different temperatures.

Under these conditions we can define the thermopower $\hat{S}$, which describes the relation between a temperature gradient and an internal electric field $\mathbf{E}$ induced by a charge accumulation

$$\mathbf{E} = \frac{1}{e} \nabla \mu = \frac{1}{2e} (\nabla \mu^+ + \nabla \mu^-) = \hat{S} \nabla T. \quad (61)$$

Similarly, it is possible to define the spin Seebeck coefficient $\hat{S}^s$ as

$$\frac{1}{2e} (\nabla \mu^+ - \nabla \mu^-) = \hat{S}^s \nabla T, \quad (62)$$

which describes the spin accumulation due to a temperature gradient. In order to derive the relevant quantities, we need to find an expression for the charge and spin accumulation as a function of the temperature gradient. The most convenient situation is the condition where both partial currents vanish. With the help of Eq. (53) we can define the spin-dependent Seebeck coefficients

$$\frac{1}{e} \nabla \mu^\pm = -\frac{1}{eT} (\hat{L}_0^\pm)^{-1} \hat{L}_1^\pm \nabla T = \hat{S}^\pm \nabla T. \quad (63)$$

Then, the thermopower and the spin Seebeck coefficient can be expressed in terms of $\hat{S}^\pm$ as

$$\hat{S} = \frac{1}{2} (\hat{S}^+ + \hat{S}^-) \quad \text{and} \quad \hat{S}^{\text{spin}} = \frac{1}{2} (\hat{S}^+ - \hat{S}^-), \quad (64)$$

respectively. In the following, we derive the elements of both tensors for nonmagnetic cubic crystals. Taking into account Eqs. (57) and (58), we obtain

$$\hat{S}^\pm = -\frac{1}{eT} \times \quad (65)$$

$$\begin{pmatrix} \frac{L_{0xx}^+ L_{1xx}^+ + L_{0yx}^+ L_{1yx}^+}{(L_{0xx}^+)^2 + (L_{0yx}^+)^2} \mp \frac{L_{0xx}^+ L_{1yx}^+ - L_{0yx}^+ L_{1xx}^+}{(L_{0xx}^+)^2 + (L_{0yx}^+)^2} & 0 \\ \pm \frac{L_{0xx}^+ L_{1yx}^+ - L_{0yx}^+ L_{1xx}^+}{(L_{0xx}^+)^2 + (L_{0yx}^+)^2} & \frac{L_{0xx}^+ L_{1xx}^+ + L_{0yx}^+ L_{1yx}^+}{(L_{0xx}^+)^2 + (L_{0yx}^+)^2} \\ 0 & 0 & \frac{L_{1zz}^+}{L_{0zz}^+} \end{pmatrix}$$

and

$$\hat{S} = \begin{pmatrix} S_{xx} & 0 & 0 \\ 0 & S_{xx} & 0 \\ 0 & 0 & S_{zz} \end{pmatrix}, \quad \hat{S}^s = \begin{pmatrix} 0 & -S_{yx}^s & 0 \\ S_{yx}^s & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad (66)$$

Spin caloric transport

according to Eq. (64). The nonvanishing elements of the thermopower and the spin Seebeck coefficient are given by

$$S_{xx} = -\frac{1}{eT} \frac{L_{0xx}^+ L_{1xx}^+ + L_{0yx}^+ L_{1yx}^+}{(L_{0xx}^+)^2 + (L_{0yx}^+)^2}, \quad S_{zz} = -\frac{1}{eT} \frac{L_{1zz}^+}{L_{0zz}^+}, \quad (67)$$

and

$$S_{yx}^{\text{spin}} = -\frac{1}{eT} \frac{L_{0xx}^+ L_{1yx}^+ - L_{0yx}^+ L_{1xx}^+}{(L_{0xx}^+)^2 + (L_{0yx}^+)^2}. \quad (68)$$

The important quantities for the considered phenomenon are S_{xx} and S_{yx}^s describing the charge and spin accumulation, respectively. Normally, the off-diagonal elements of L_m^+ are much smaller than the corresponding diagonal elements. Therefore, we may introduce the approximate expression for the thermopower

$$S_{xx} \approx S_{xx}^0 = -\frac{1}{eT} \frac{L_{1xx}^+}{L_{0xx}^+}. \quad (69)$$

To evaluate the strength of the SNE for the case considered here the spin Nernst angle can be defined as [53]

$$\theta_{\text{SN}} = S_{yx}^{\text{spin}} / S_{xx}. \quad (70)$$

The corresponding results obtained for several Cu-based dilute binary alloys are shown in Sec. IV.

5.3.2. Spin current In the following, we consider another situation, where we allow the spin current to flow instead of the creation of the spin accumulation, which means $j_y^s \neq 0$ and $\nabla_y \mu^+ - \nabla_y \mu^- = 0$. The system is still electrically insulated in the x direction with no macroscopic currents flowing along this direction leading to

$$L_{0,xx}^\pm \nabla_x \mu^\pm + \frac{1}{T} L_{1,xx}^\pm \nabla_x T = 0 \quad (71)$$

and

$$(\mathcal{J})_y^\pm = -L_{0,yx}^\pm \nabla_x \mu^\pm - \frac{1}{T} L_{1,yx}^\pm \nabla_x T \quad (72)$$

which using Eqs. (71) and (58) gives us

$$L_{0,xx}^+ (\nabla_x \mu^+ + \nabla_x \mu^-) = -\frac{2}{T} L_{1,xx}^+ \nabla_x T. \quad (73)$$

Combining this with Eq. (62) it provides the thermopower

$$S_{xx}^0 = -\frac{1}{eT} \frac{L_{1xx}^+}{L_{0xx}^+}, \quad (74)$$

describing the Seebeck effect in the short-circuit case in y direction, as indicated by the introduced superscript 0 pointing to the fact that this expression is valid for a vanishing spin accumulation in the transverse direction. Since a finite spin accumulation is caused otherwise by the spin-orbit coupling, Eq. (74)

Figure 11. (Color online) Compensation of the thermal current by the induced electrical current. Here $T_1 < T_2$ is assumed.

coincides with the thermopower obtained within the nonrelativistic treatment. [1]

In order to define a physical quantity to measure the strength of the SNE, the analogy to the spin Hall conductivity can be used. The latter case provides a linear relation between the spin current density and the applied electric field. [16] Similarly, for the SNE we relate the transverse spin current density j_y^s to the temperature gradient via the so-called spin Nernst conductivity (SNC) [53]

$$(\mathcal{J}^s)_y = \sigma_{\text{SN}} \nabla_x T. \quad (75)$$

In Ref. [96] this quantity was called the thermospin Hall conductivity, however the name *spin Nernst conductivity* fits the considered phenomenon better. It is also worth mentioning that this quantity does not have the conventional units of charge or spin conductivities but is measured in $\text{A}/(\text{K} \cdot \text{m})$.

Taking into account Eqs. (72) and (58) we write

$$(\mathcal{J}^s)_y = -L_{0,yx}^+ (\nabla_x \mu^+ + \nabla_x \mu^-) - \frac{2}{T} L_{1,yx}^+ \nabla_x T \quad (76)$$

for the y -component of the spin current defined by Eq. (52). With Eqs. (74), (75), and (76) we obtain the following expression for the spin Nernst conductivity

$$\sigma_{\text{SN}} = -2eS_{xx}^0 L_{0yx}^+ - \frac{2}{T} L_{1yx}^+. \quad (77)$$

Combining this with Eq. (74) we find

$$\sigma_{\text{SN}} = \sigma_{\text{SN}}^E + \sigma_{\text{SN}}^T = \frac{2}{T} \frac{L_{1xx}^+}{L_{0xx}^+} L_{0yx}^+ - \frac{2}{T} L_{1yx}^+, \quad (78)$$

where the two separate terms provide the contributions either directly from the temperature gradient (σ_{SN}^T) or via the Seebeck effect (σ_{SN}^E).

A simplified picture for an illustration of this result is given by Fig. 11, showing two macroscopic electrical currents which compensate each other. The first of them, $\mathcal{J}_T$, is related to the thermal current due to the applied temperature gradient. The second contribution, $\mathcal{J}_E$, arises from the Seebeck effect. Together they give no charge current along x direction,

Table 2. The spin Nernst angle θ_{SN} (dimensionless) and the efficiency γ (in $10^{-3} \times 1/V$) for the Cu(Au), Cu(Ti), Cu(Bi), and Cu(Zn) alloys. Both quantities are obtained at 300 K.

Alloy	Cu(Au)	Cu(Ti)	Cu(Bi)	Cu(Zn)
θ_{SN}	-0.26	0.0046	-0.027	0.0069
γ	-6.54	3.67	5.19	-2.13

$\mathcal{J}_T + \mathcal{J}_E = 0$, but each of them can provide a different amount of transverse spin current. A similar macroscopic explanation was given in Ref. [96] but using another language. Namely, let us assume that $T_2 > T_1$ in Fig. 11. The absence of the electrical current means that the number of “hot” electrons, coming from the right to the left side of the sample, is one and the same as the number of “cold” electrons, which come from the left to the right side. Without the temperature gradient ($T_2 = T_1$) they would provide the same amount of spin currents flowing in opposite directions and consequently canceling each other. An applied temperature gradient leads to $T_2 \neq T_1$ and consequently the two parts of the oppositely moving electrons have different transport properties. Therefore, they provide unequal amounts of opposite spin currents, resulting in a finite net spin current. Within this picture the origin of the transverse spin current is attributed to the heat current induced by the temperature gradient, which makes the analogy to the spin Hall effect not so obvious.

In addition to these simplified macroscopic pictures, it is possible to provide a more detailed explanation of the SNE by considering the nonequilibrium microscopic currents

$$\mathcal{J}_k = -\frac{e}{V} \frac{\partial f_k^0}{\partial E_k} (\mathbf{v}_k \circ \mathbf{\Lambda}_k) \left[\nabla \mu + \frac{E_k - \mu}{T} \nabla T \right], \quad (79)$$

as follows from Eq. (46). Although the corresponding macroscopic charge currents $\mathcal{J} = \sum_k \mathcal{J}_k = 0$ vanish, the separate microscopic currents remain nonzero. They cause corresponding spin currents via the spin Hall effect occurring at a microscopic level which results in a finite macroscopic spin current. Such a microscopic consideration helps to understand the analogy of the considered phenomenon to the SHE.

In order to describe the efficiency of the SNE for the considered situation, we use the ratio of the spin Nernst current given by Eq. (75) and the heat current $(\mathcal{Q})_x = -\kappa \nabla_x T$, [117]

$$\gamma = \frac{\sigma_{\text{SN}}}{-\kappa}, \quad (80)$$

where κ is the heat conductivity [1, 117]

$$\kappa = \frac{2}{eT} \left(\frac{(L_{1xx}^+)^2}{L_{0xx}^+} - L_{2xx}^+ \right). \quad (81)$$

The corresponding results are presented in the following section.

5.4. Results

In this section we present the results for Au, Bi, Ti, and Zn substitutional impurities in Cu. The electronic structure of the Cu host was obtained by means of a fully relativistic Korringa-Kohn-Rostoker method. [115] The impurity problem was solved self-consistently in real space on a cluster of 55 atoms, including four nearest neighbor shells for charge relaxation. In order to maintain a balance between reliability and computational efforts, we evaluated Eq. (50) equidistantly at 25 energies $E_F - 0.024 \text{ Ry} < E < E_F + 0.024 \text{ Ry}$ around the Fermi level. This is facilitated by applying a Gaussian smoothing followed by a cubic spline interpolation for the computation of Eq. (49). A detailed description of the procedure is given in Appendix.

Table 2 shows the strength of the SNE for the open-circuit and short-circuit cases described by θ_{SN} and γ , respectively. Both quantities are practically temperature independent. Therefore, their values are shown at 300 K only. There is no evident correlation between the creation of the spin accumulation or the spin current, neither with respect to the magnitude nor the sign of θ_{SN} and γ . Nevertheless, the strongest SNE is provided by the Cu(Au) alloy in both cases. In order to get more insight into the underlying physics, in Figure 12 we consider constituents of Eqs. (70) and (80). With our choice of the impurities in a Cu host, all four possible combinations with respect to the sign of the thermopower and the spin Seebeck coefficient are obtained. Whereas the thermopower and spin Seebeck coefficient are positive in the entire investigated temperature range for Ti impurities in Cu, both are negative for the Cu(Zn) alloy. By contrast, one of these quantities is positive whereas the other is negative for Au or Bi impurities, respectively. The final graph shows the SNC in comparison to the heat conductivity, which helps to understand the results of Table 2. Indeed, now it is possible to reveal that actually σ_{SN} and S_{yx}^s have opposite sign. However, the heat conductivity is a positive-defined quantity, whereas the thermopower can have arbitrary sign. Consequently, the correlation between θ_{SN} and γ is absent.

The results for the two contributions to the total spin Nernst conductivity, as given by Eq. (78), are shown in Fig. 13. Here, for the Cu(Ti) and Cu(Bi) alloys σ_{SN}^E and σ_{SN}^T have opposite sign. Consequently, they compensate each other partially. For the Cu(Zn) alloy both contributions are positive and of the same order of magnitude facilitating the effect. As was mentioned already above, the largest SNC is obtained

Figure 12. (Color online) The thermopower S_{xx} , the spin Seebeck coefficient S_{yx}^s , the spin Nernst conductivity σ_{SN} , and the heat conductivity κ for four different substitutional impurities in a Cu host at an impurity concentration 1 at. %.

Figure 13. (Color online) The spin Nernst conductivity for four different substitutional impurities in a Cu host at an impurity concentration 1 at. %.

for the Cu(Au) alloy. However, now one can see that it is dominantly provided by $\sigma_{\text{SN}}^{\text{T}}$, which is caused by the corresponding large charge conductivity. [53]

As shown in Fig. 13, the quantities S_{xx} , S_{yx}^{spin} , σ_{SN} , and κ have a nearly linear behavior with respect to the temperature despite the prefactor $1/T$. This comes from the fact that the conductivity is approximately

linear in energy for the relevant energy interval. Let us assume

$$\hat{\sigma}(E) \approx \hat{\sigma}(\mu) + (E - \mu) \left. \frac{d\hat{\sigma}(E)}{dE} \right|_{E=\mu} \quad (82)$$

and use Eq. (49), then due to symmetry the second and first terms in Eq. (82) vanish for $n = 0$ and $n = 1$,

Figure 14. (Color online) The charge conductivity σ_{xx} , the spin Hall conductivity σ_{yx}^s , the thermopower S_{xx} , the spin Seebeck coefficient S_{yx}^s , the spin Nernst conductivity σ_{SN} , and the heat conductivity κ as obtained either neglecting (blue/gray) or including (red/black) spin-flip processes. The actual values are divided by the corresponding quantity obtained by incorporating spin flipping.

respectively. Consequently, we find

$$\hat{L}_0(T) \approx -\frac{1}{e} \int dE \hat{\sigma}(\mu) \left(-\frac{\partial f_0}{\partial E} \right) \approx -\frac{1}{e} \hat{\sigma}(\mu), \quad (83)$$

which implies the quantity $\hat{L}_0$ gives only a constant factor. By contrast, according to Ziman, [1] the quantities

$$\begin{aligned} \hat{L}_1(T) &\approx -\frac{1}{e} \int dE (E - \mu)^2 \frac{d\hat{\sigma}(E)}{dE} \bigg|_{E=\mu} \left(-\frac{\partial f_0}{\partial E} \right) \\ &\approx -\frac{1}{e} \frac{\pi^2}{3} (k_B T)^2 \frac{d\hat{\sigma}(E)}{dE} \bigg|_{E=\mu} \end{aligned} \quad (84)$$

and

$$\begin{aligned} \hat{L}_2(T) &\approx -\frac{1}{e} \int dE (E - \mu)^2 \hat{\sigma}(\mu) \left(-\frac{\partial f_0}{\partial E} \right) \\ &\approx -\frac{1}{e} \frac{\pi^2}{3} (k_B T)^2 \hat{\sigma}(\mu) \end{aligned} \quad (85)$$

have a quadratic dependence on temperature. This explains the linear behaviour for S_{xx} , S_{yx}^{spin} , σ_{SN} and κ .

An important point is the influence of spin-flip scattering on the considered transport properties. In Ref. [53] the spin-flip processes were neglected in the calculations. Figure 14 illustrates their influence on the different transport properties. Since the impact of spin-flip scattering for Ti or Zn impurities in Cu is

negligible, we restrict our consideration to the Cu(Au) and Cu(Bi) alloys, where the effect is much stronger. Despite the fact that bismuth is a bit heavier than gold, spin flipping has stronger influence for Cu(Au) than for the Cu(Bi) alloy. As expected from a general point of view, the transverse components (σ_{yx}^s , S_{yx}^{spin} , σ_{SN}) are more affected by spin relaxation than the longitudinal transport properties.

5.5. Conclusion

A detailed consideration of the first-principles semiclassical approach for description of the skew-scattering contribution to the spin Nernst effect is provided. Both, open and closed boundary conditions, leading to either spin accumulation or spin currents transverse to an applied temperature gradient, are studied intensively. The solution of the linearized Boltzmann equation, required for a practical implementation of the proposed formalism, is realized by means of the relativistic Korringa-Kohn-Rostoker method. Using the examples of dilute binary alloys composed of Cu with Au, Bi, Ti, or Zn impurities, we present the results for different transport properties related to the considered phenomenon. It is shown that their strength and sign can be well tuned by means of the type of substitutional impurities, which provides an opportunity to design practical devices with defined characteristics. In addition, we reveal that the spin-flip scattering can have strong influence on the spin Nernst effect, especially via the change of the corresponding transverse transport properties.

5.6. Appendix

The computation of the transport coefficients according to Eqs. (49) and (50) involves two integrals that need to be evaluated carefully. First, the integral of Eq. (49) requires an energy span which is large enough to provide the specified accuracy

$$\int_{E_{\min}}^{E_{\max}} \left(-\frac{\partial f^0(E, T)}{\partial E} \right) dE > 0.9999 \quad (86)$$

for the highest chosen temperature (300 K) and dense enough to ensure an accurate numerical integration for the lowest chosen temperature (30 K)

$$\begin{aligned} \int dE \hat{\sigma}(E) \left(-\frac{\partial f^0(E, T)}{\partial E} \right) (E - \mu)^m = \\ \sum_{E_i} \Delta E_i \hat{\sigma}(E_i) \left(-\frac{\partial f^0(E, T)}{\partial E} \right) \bigg|_{E=E_i} (E_i - \mu)^m. \end{aligned} \quad (87)$$

Second, to calculate the conductivity given by Eq. (50), one needs to evaluate the Fermi surface integral which strongly depends on its sample quality.

As shown by Fig. 15, a proper description of the energy-dependent conductivity requires a huge number of elements (triangles and quadrangles) for the Fermi surface sampling. The straightforward approach requires a few hundred thousands surface elements for the relativistic irreducible part of the Brillouin zone, which makes the computations extremely demanding and almost not feasible even for such a simple case as the Cu host. However, the problem can be avoided by smoothing the conductivity

$$\hat{\sigma}(E_j) = \frac{\sum_i g_i(E_j) \hat{\sigma}(E_i)}{\sum_i g_i(E_j)} \quad (88)$$

at the energy E_j using the Gaussian

$$g_i(E_j) = \exp \left[- \left(\frac{E_i - E_j}{b} \right)^2 \right] \quad (89)$$

with the parameter $b = 0.0034$ Ry found to provide an optimal procedure. Based on 25 equidistant energy calculations within $E_F - 0.024$ Ry $< E_i < E_F + 0.024$ Ry, we evaluated 21 equidistant smoothed values in the range $E_{\min} = E_F - 0.02$ Ry $< E_j < E_F + 0.02$ Ry $= E_{\max}$ which turned out to be sufficient to fulfill the condition of Eq. (86). In order to satisfy the requirement of Eq. (87), we interpolated the 21 values by means of cubic splines on a very dense energy mesh allowing for reliable numerical integration. The result of this procedure is shown as the purple solid graph in Fig. 15. Requiring much less computational efforts, it gives the same energy dependence as the setup using a Fermi surface with nearly four times as many surface elements.

Figure 15. (Color online) The energy-dependent charge conductivity of the Cu(Au) alloy around the Fermi level calculated for four different setups neglecting spin-flip processes. The relativistic irreducible part of the Brillouin zone was filled with different numbers of equally distributed $\mathbf{k}$ points leading to 7926 (orange circles), 31458 (green triangles), or 125184 (blue squares) surface elements of the Fermi surface in this part. The solid purple line shows the results based on 31458 surface elements post-processed by the Gaussian smoothing and cubic spline interpolation.

6. *Ab-initio* modelling of spin-caloric transport in nanostructured Heusler alloys

Some ferromagnetic intermetallic compounds of two transition metals and one main group element display remarkable electronic properties due to short-range atomic order. Within the scalar-relativistic approximation, some of them are ferromagnetic half-metals. This statement implies a finite electronic density of states for one spin channel, but a gap around the Fermi energy in the electronic band structure of the other spin channel. Consequently, such materials are seen as the most promising candidates for being incorporated in spin filters and spin injectors devices, including their thermally driven counterparts [3, 118].

Heusler alloys are ternary intermetallic compounds with the cubic $L2_1$ structure. The cobalt-derived compounds, in particular Co_2TiZ and Co_2MnZ (Z being one of the group-IV elements Si, Ge or Sn) are ferromagnets that meet the above-mentioned criteria. Of these, alloys containing Ti have experimentally been shown to display a high thermopower with negative sign [119, 120]. Motivated by this experimental finding, we have focused on layered nanostructures containing these materials for studying their spin-caloric transport properties.

First, we establish the physics of thermopower in the bulk materials Co_2TiSi , Co_2TiGe and Co_2TiSn and investigate the sensitivity of the Seebeck coefficient to intrinsic point defects [121]. In these Heusler alloys the Ti atom has a negligible magnetic moment, while the Co atoms have itinerant moments of $1 \mu_B$. In the second part of this section, we analyze the spin-dependent Seebeck coefficient of an envisioned spin injection device that consists of a few-atomic-layer insertion of Co_2TiSi or Co_2TiGe between aluminium leads [122]. A comparable study has been performed for heterostructures of platinum leads [123], motivated by the observation that Pt is most commonly used as detector for spin accumulation via the inverse spin Hall effect. In this study, the focus was on spacer layers consisting of ferromagnetic Heusler alloys with high Curie temperatures, such as Co_2FeSi , Co_2FeAl , Co_2MnSi and Co_2MnAl .

For calculating the formation energy of intrinsic point defects in Co_2TiZ , we used a supercell method in conjunction with a DFT calculation within a plane-wave electronic structure code [124]. The results have shown that the defects with the lowest formation energy are Co vacancies (V_{Co}) and Ti_{Sn} anti-sites [121]. A thermodynamic analysis based on the calculated formation energy reveals that their formation may occur spontaneously under Co-poor preparation conditions, indicated by a negative value of the formation energy in a part of the thermodynamically allowed interval of chemical

potentials.

In a second step, we performed electronic transport calculations of selected defected structures. The point defects were described employing the coherent potential approximation (CPA) within the Korringa-Kohn-Rostoker Green function method in its spin-polarized relativistic implementation (SPR-KKR) [35, 33, 125]. Subsequent to the SCF calculations, the temperature-dependent longitudinal Seebeck coefficient $S_{ii}(T)$ (with $i = x, y$, or z the Cartesian coordinate) was obtained from the diagonal elements of the energy dependent conductivity tensor $\sigma_{ii}(E)$ [21], determined via the Kubo-Greenwood formula. Introducing the transport coefficients

$$L_{m,ij} = -\frac{1}{e} \int \sigma_{ij}(E) \left[-\frac{\partial f^0(E, \mu, T)}{\partial E} \right] (E - \mu)^m dE, \quad (90)$$

where $f^0(E, \mu, T)$ is the Fermi-Dirac distribution function with chemical potential μ at energy E for the temperature T , the Seebeck coefficient is given by

$$S_{ii}(T) = -\frac{1}{eT} \frac{L_{1,ii}}{L_{0,ii}} \quad (91)$$

with e the elementary charge. The adopted approach enabled us to study the effect of the defects on both conductivity and thermopower in the limit of fairly low defect concentration, appropriately accounting for the vertex corrections to the current-current correlation function [36].

The main goal of our investigations was to establish a connection between the different features of the Seebeck coefficient seen in Fig. 17(b) with the defect-induced modifications in the electronic structure. In particular, we found that the Seebeck coefficient in the presence of point defects is very sensitive to details of the electronic structure, and relativistic effects (spin-orbit coupling) must be taken into account. Indeed, in a fully relativistic treatment, spin is no longer a good quantum number and a projection of the density of states onto spin channels will indicate a small density of states even in the region of the half-metallic gap [126]. The calculated spin-polarized relativistic dispersion relation shown in Fig. 16 for the perfectly stoichiometric Co_2TiSi Heusler alloy shows that the strong spin mixing occurring in the proximity of the Fermi energy E_F is the result of several band anti-crossings (evidenced by the highlighted regions) between a Co majority-d band crossing E_F and a rather flat Co minority-d band lying $0.2 - 0.4$ eV above.

We mainly focused on those defects that have low formation energy, namely the Co vacancies (V_{Co}) and the Ti_{Sn} as well as the Co_Z or Co_{Ti} anti-sites. Based on our thermodynamic analysis it must be expected that they can be formed unintentionally during materials

Figure 16. Spin-polarized relativistic dispersion relation E_k along selected high symmetry directions in the fcc BZ, calculated for the Co_2TiSi full Heusler alloy in the $L2_1$ structure obtained using the FP-SPR-KKR method. The energy is given relative to the Fermi energy E_F of the system. The highlighted areas located $0.2 - 0.4$ eV above E_F along the $W - \Gamma$, $K - \Gamma$, and $\Gamma - X$ directions point to anti-crossings between a Co majority-d and a Co minority-d band, which occur as a result of spin-orbit coupling.

synthesis. Remarkably, a negative $S(T)$ is obtained for Co_2Z and VcCo defects due to subtle changes of the electronic structure mediated by spin-orbit coupling (SOC). In these two systems, minority-spin defect-induced states appear right above the Fermi energy hybridizing with the minority-spin states shown in Fig. 16. As a result of their coupling to the majority-spin states crossing E_F , the otherwise rather localized impurity states *gain mobility* and cause an enhanced conductivity above E_F , with the net result of a strongly positive asymmetric moment L_1 in Eq. (91), and thus to a large, negative Seebeck coefficient. As such, the assumption that real samples contain some finite concentration of these defects is in line with the experimentally observed qualitative features for $S(T)$ [119, 120].

The calculated energy-dependent electronic conductivity of the $\text{Co}_2\text{TiSi:D}$ dilute alloys is shown in Fig. 17(a). We only plot the longitudinal $\sigma_{xx}(E)$, given relative to the Fermi energy E_F of each system (dashed vertical line). While a smooth dependence of all $\sigma_{xx}(E)$ curves in the $E_F \pm 0.15$ eV energy interval is obvious, several peculiarities can be noted: (i) in four cases the conductivity dependence on its energy argument exhibits a negative slope around E_F ; (ii) a steep increase of $\sigma_{xx}(E)$ occurs at energies above $\simeq 0.18$ eV; (iii) in two of the investigated systems, $\text{Co}_2\text{TiSi:VcCo}$ and $\text{Co}_2\text{TiSi:CoSi}$, the conductivity is characterized by a positive slope around the Fermi energy.

By virtue of Eqs. (90) and (91), the Seebeck coefficient $S(T)$ is expressed as the quotient of the first

and zeroth moments of the conductivity. The findings related to the changes in the energy dependence of $\sigma_{xx}(E)$ can then be directly transferred to the corresponding results obtained for $S(T)$ presented above in Fig. 18. If we made, in the absence of better knowledge, the assumption of energy-independent scattering, our theoretical results would predict a positive Seebeck coefficient for the bulk Co_2TiZ materials [119, 122], a finding which is in contrast to the large, negative values reported experimentally, reaching as much as $-30\mu\text{V/K}$ in $\text{Co}_2\text{TiSi/Ge}$ and $-50\mu\text{V/K}$ in Co_2TiSn [119, 120]. Calculating the conductivity via the Kubo-Greenwood formula, we are not limited to such an assumption; rather, the scattering properties of the point defects are taken into account within the framework of the CPA. As seen in Figs. 17 and 18, the tendency of a positive Seebeck coefficient is indeed retained for the systems that have a weakly energy-dependent scattering, such as CoTi , TiCo , SiTi , and TiSi . As the temperature increases, the contribution of the minority-spin electrons becomes more important. This is because the minority-spin bands above the half-metallic gap become populated at higher temperatures. As shown above, in the case of defects with a weakly energy-dependent scattering the onset of conductivity in the minority-spin bands leads to an abrupt rise in the conductivity and thus to a negative, electron-like contribution to the Seebeck coefficient. As a result, the Seebeck coefficient drops sharply and becomes negative at elevated temperatures for the systems containing CoTi , SiTi and TiSi . One notes here the direct relation between the change of slope for $S(T)$ and the ever increasing contribution coming from the flat minority-spin band above the half-metallic gap. The exception to this behaviour is represented by the TiCo anti-site where the presence of a impurity band below E_F was shown to add a hole-like contribution to the conductivity. This contribution turns out large enough such as to preserve a positive Seebeck coefficient throughout the whole investigated temperature range.

In some systems, however, the scattering of electrons by point defects carrying a magnetic moment has a more profound effect on the electronic structure and even leads to a mixing of electronic bands with different prevailing spin character. In systems with a strongly energy-dependent scattering, the conductivity was found to be larger above E_F than below. This leads to a negative Seebeck coefficient, as obtained for $\text{Co}_2\text{TiSi:VcCo}$ and $\text{Co}_2\text{TiSi:CoSi}$, see Fig. 17. We note that only in these cases the results qualitatively reproduce the experimentally observed behaviour. Quantitatively, the calculated $S(T)$ for the CoSi anti-site deviates within 30 % from the experimental data for $T \leq 300$ K.

Figure 17. Calculated transport properties for several off-stoichiometric native defects in Co_2TiSi , modelled as dilute alloys with 3 % concentration: (a) the energy dependence of the diagonal element $\sigma_{xx}(E)$ of the electronic conductivity tensor and (b) the temperature-dependent Seebeck coefficient $S(T)$.

Analogous considerations can be applied when analyzing the dependence of the Seebeck coefficient on the group IV element Z in the isoelectronic systems Co_2TiZ : D . The corresponding results for all investigated defects at a net composition of $x = 0.03$ are shown in Fig. 18. Although the conductivity is mainly dominated by majority-spin carriers, one can ascribe the various differences in the Z -atom dependence of $S(T)$ as being mostly related to changes occurring in the minority-spin channel. We have previously identified three systems, Co_{Ti} , Ti_Z , and Z_{Ti} , which are characterized by a sudden change of slope in $S(T)$ at elevated temperatures. This behaviour was associated with the minority-spin bands above the half-metallic gap becoming populated with increasing temperature. As the offset of these bands relative to E_F increases with Z , the peak in $S(T)$ also shifts towards higher T values when Z changes from Si to Ge and then Sn.

For the two defects for which the Seebeck coefficient had a negative sign, $Co_2TiSi:Vc_{Co}$ and $Co_2TiSi:Co_{Si}$, we find this sign to persist for Co_2TiGe and Co_2TiSn . Moreover, we note that for both defects $|S(T)|$ increases with increasing atomic number of the group IV element Z and therefore with the strength of the spin-orbit interaction. This is a very important observation: as argued above, it is the SOC that, by mixing the defect-induced states with the highly dispersive majority-spin bands crossing E_F increases the transmission above the Fermi level and thus the asymmetry term $L^{(1)}$, the numerator of Eq. (91).

Having analyzed the properties of realistic bulk Heusler alloys (including point defects and off-stoichiometry), we now turn to their applications in nanostructures. As a prototypical system of practical interest, we have studied atomically thin layers of Co_2TiSi and Co_2TiGe embedded between aluminium

electrodes.

Since the spin relaxation length of Al is particularly large, Al leads lend themselves as conductors for a spin-polarized current that is induced by applying a temperature gradient perpendicular to the layers. Moreover, the crystal lattice of face-centred cubic Al can be matched, after rotated by 45° , to the cubic lattices of Co_2TiSi and Co_2TiGe , whereas the lattice constant of Co_2TiSn is somewhat too large. In the first two systems, the mismatch strain is accommodated by a tetragonal distortion of the Heusler structure, which was taken into account in the calculations by fully relaxing the epitaxial heterostructures. Since we restrict ourselves to nano-contacts and very thin layers, the likelihood of incorporating point defects in this very small amount of material of the Heusler layer can be considered small. In the focus of our study was the issue of half-metallicity being preserved in very thin layers, and the role of interfaces between Al and the Heusler alloys on the spin-caloric transport properties. For this purpose we considered two idealized interfaces, one where the Heusler alloy is terminated by a complete Co layer in contact with Al, and another one where a mixed layer of the Heusler structure (Ti-Si or Ti-Ge, depending on the chosen alloy) is in contact with Al. Our calculation of the interface energies [122] have shown that the Co interface is stable under equilibrium conditions, whereas the TiSi or TiGe interface are less stable, but can be prepared under non-equilibrium conditions.

The question of half-metallicity being retained in the thin films is best studied in a scalar relativistic approach to the electronic structure. In such an approach, the minority-spin channel in the bulk materials displays a gap around the Fermi level. Comparing the layer-resolved electronic density of states of the heterostructures with those of ideal bulk

Figure 18. The Seebeck coefficient $S(T)$ calculated for selected defects in $\text{Co}_2\text{TiZ:D}$ at 3 % defect composition. In each panel, the curves corresponding to different group IV elements Z are represented by the (red) thin solid, (green) long dashed, and (blue) thick solid lines for Si, Ge, and Sn, respectively.

structures allows us to identify possible metal-induced gap states. The calculations have shown that the hybridization of electronic states of Al with those of the Heusler material occurs in particular for the Co-termination, for which the interfacial bond length is shorter than for the TiSi or TiGe termination. Even in very thin films, however, one finds that the half-metallic gap is recovered in the central layers of the Heusler alloy, already 3 to 4 atomic planes away from the interface [122].

The conductance of Al/ Co_2TiZ /Al heterostructures perpendicular to the planes is governed by the backscattering of carriers from the interfaces between Al and the Heusler alloy. Thus, a suitable approach to the conductance is provided by the Landauer-Büttiker formula generalized to spin-polarized systems. From the accurately converged self-consistent DFT potentials of the leads and the scattering regions, transport coefficients are calculated separately for both spin channels using the method described in Refs. [127] and [128]. Starting from the energy- and spin-resolved transmission probability $\Gamma^\sigma(E)$,

$$\Gamma^\sigma(E) = \frac{1}{A_{\text{BZ}}} \int d\mathbf{k}_{\parallel} \Gamma^\sigma(\mathbf{k}_{\parallel}, E), \quad (92)$$

the Seebeck coefficients are evaluated using the approach of Sivan and Imry[129] starting from the central quantity $\Gamma^\sigma(E)$ and the Fermi occupation function $f^0(E, T, \mu)$. Within this formalism, employing Mott's two-current model, the spin-projected conductance is expressed as

$$G^\sigma(T) = \frac{e^2}{h} \int dE \Gamma^\sigma(E) \left[-\frac{\partial f^0}{\partial E} \right]. \quad (93)$$

while the spin-projected Seebeck coefficient takes on

Figure 19. (a) Effective, (b) spin-dependent, and (c) spin-projected Seebeck coefficients calculated for the Al/ Co_2TiSi /Al and Al/ Co_2TiGe /Al trilayer systems. The labelling of the curves designates the respective spacer material and the Al-Heusler interface termination (Co-Co or Ti-Z) in parentheses.

the form

$$S^\sigma(T) = -\frac{1}{eT} \frac{\int dE \Gamma^\sigma(E) \frac{\partial f^0}{\partial E} (E - E_F)}{\int dE \Gamma^\sigma(E) \frac{\partial f^0}{\partial E}}, \quad (94)$$

formulas which are completely analogous to Eqs. (90)-(91) but use the transmission probability rather than

the electronic conductivity [130].

Using the two quantities above, we can derive the effective Seebeck coefficient

$$S = \frac{G^\uparrow S^\uparrow + G^\downarrow S^\downarrow}{G^\uparrow + G^\downarrow}, \quad (95)$$

as well as the spin-dependent Seebeck coefficient

$$S^{\text{spin}} = \frac{G^\uparrow S^\uparrow - G^\downarrow S^\downarrow}{G^\uparrow + G^\downarrow}. \quad (96)$$

The latter quantity describes the proportionality factor between the temperature difference applied at the leads and the spin accumulation (i.e., the difference in chemical potential for spin-up and spin-down electrons) that can be reached under open-circuit conditions. In a closed electronic circuit, the temperature gradient will then induce a spin-polarized stationary current.

The calculated effective and spin-dependent Seebeck coefficients for the two systems Al/Co₂TiSi/Al and Al/Co₂TiGe/Al with different terminations are shown in a compact form in Fig. 19(a) and (b), respectively, for temperatures up to 350 K. Each $S(T)$ curve is labelled according to the convention “spacer material (termination)”.

Equations (95) and (96) express the effective and spin-dependent Seebeck coefficients as a weighted sum (difference) of the auxiliary quantities S^σ , defined by Eq. (94), which are not accessible to direct electrical measurement. In the above analysis, the calculated $S^\uparrow(T)$ (majority-spin) and $S^\downarrow(T)$ (minority-spin) can be considered in analogy to parallel connected resistors.

As can be seen from the first row of Fig. 19, the magnitude of both S and S^σ follows a sequence in which the systems with a mixed interface layer (Ti-Si or Ti-Ge) show positive values, while the Al/Co₂TiGe/Al system with Co-Co terminated interfaces shows negative values. The Al/Co₂TiSi/Al system with Co-Co terminated interfaces is somewhat peculiar because of its very small values of both the effective as well as spin-dependent Seebeck coefficient $S^{\text{spin}}(T)$.

Comparing panels (a) and (b) to (c) and (d) in Fig. 19, it becomes clear that S is mainly determined by the majority-spin component $S^\uparrow$: the same sequence from positive to negative values in the temperature dependence appears in both $S^\uparrow$ and in S . The minority-spin component $S^\downarrow$, in turn, exhibits a fairly similar T -dependence regardless of spacer and termination. One recalls that a band gap is present in the minority-spin channel, rendering the spin-down transmission into a “tunnelling through a semiconductor”-equivalent case. Since $S^\downarrow$ is weighted with the minority-spin conductance $G^\downarrow$ in Eqs. (95) and (96), this explains why its contribution to both S and S^{spin} , in spite of the very large values, is drastically diminished.

In order to rationalize the above findings, it is essential to disentangle the contributions of electrons with various momenta parallel to the interfacial plane to the overall transmission probability, i.e., the contributions of $\Gamma^\sigma(\mathbf{k}_\parallel, E)$ in different regions of the Brillouin zone (BZ) to the integral in Eq. (92). We show in Fig. 20 contour plots of the majority-spin transmission probability $\Gamma^\uparrow(\mathbf{k}_\parallel, E)$ for $E = E_F$, in the full 2D-BZ and for all combinations spacer material plus interface termination studied here.

A similar pattern is found for the two Ti-Z terminations (left column): the different spacer materials only change the transmission amplitude, without introducing or removing any individual transmission channels. This is directly reflected in the Seebeck coefficients calculated for the Ti-Z terminations [cf. Fig. 19(c)] that were found to share the same qualitative energy and temperature dependence. This picture is severely altered for the Co-Co terminated Al/Co₂TiZ/Al trilayers, right panels of Fig. 20. While the transmission at E_F for the Co₂TiGe spacer shows large contributions near $\mathbf{k}_\parallel = \bar{\Gamma}$ (corresponding to normal incidence), these channels are almost blocked for Co₂TiSi (the dark blue area around $\bar{\Gamma}$). In turn, Co₂TiSi favours transmission channels along the diagonals of the 2D-BZ far from $\bar{\Gamma}$ which appear very weak in Co₂TiGe. The absence of these transmission channels around the zone centre is responsible for $\Gamma^\uparrow(E)$ being an increasing function of energy [122], and hence for the negative value of $S^\uparrow(T)$, $S(T)$ and $S^{\text{spin}}(T)$ for Co₂TiGe/(Co-Co) evident from Fig. 19. Only close to room temperature $S^{\text{spin}}(T)$ shows non-monotonic behaviour due to the increasingly important contribution from minority-spin electrons that start to bridge the half-metallic gap as the temperature is increased.

Concluding this Section, our calculations support the expectation that very thin layers of Heusler alloys that are half-metallic ferromagnets, when inserted between metallic leads, can give rise to detectable spin-caloric effects. For the usage of Al/Co₂TiZ/Al trilayers as thermally driven spin injectors, the atomic structure of the interface between Heusler alloy and Al lead turns out to be very important. According to our calculations, the largest spin accumulation is to be expected for mixed termination by Ti-Si or Ti-Ge planes of the respective Heusler material. For all materials combinations studied, including both Al and Pt leads and Heusler alloys containing Ti, Fe or Mn, the half-metallic gap was found to be recovered for Heusler films thicker than just a few lattice constants. Moreover, the relative position of the Fermi level in the lead material and the Heusler alloy is an important factor governing the relative contribution of minority- and majority-spin

Figure 20. Contour plots of the majority-spin $k_{||}$ -resolved transmission probability in Al/Co₂TiZ/Al as calculated for the energy argument $E = E_F$. Left (right) columns correspond to the Ti-Z- (Co-Co-) terminated Al/Co₂TiZ interface. The BZ centre is in the middle of each square, as indicated by the Γ point in the third panel. The k_x (k_y) axis has horizontal (vertical) orientation.

carriers. For exploiting the possibly large contribution of minority-spin electrons to the Seebeck coefficient in the semiconductor-like band structure of ferromagnetic half-metals, it would be required to bring the Fermi level close to the flat minority-spin band above the half metallic gap. This could be achieved by 'interface engineering', e.g., by a suitable electric dipole at the interface, or, in case of thicker spacer layers, by appropriately doping the Heusler alloy. For thicker Heusler films approaching bulk, the occurrence of point defects (vacancies, anti-sites) during the materials fabrication process has to be expected on the basis of formation energy calculations. We could show that the Seebeck coefficient is highly sensitive to the presence of such point defects, and hence can vary in a wide range. At elevated temperatures, scattering of electrons by magnons and/or phonons is another factor affecting the Seebeck effect under realistic measurements conditions. These aspects could make the subject of future research.

7. Tunneling Magneto Seebeck effect and thermal spin-transfer torque

7.1. Introduction

Magnetic tunnel junctions (MTJs) are a versatile tool in spintronics with a number of applications [131, 132, 133]. They are used as read sensor in hard disks, storage element in magnetic memories, and as spin resonant oscillators. Two well known effects are the tunneling magneto resistance (TMR) effect [134, 135, 136, 137, 138, 139, 140] and the spin-transfer torque (STT) [141, 142, 143, 144, 145, 146, 147, 148].

A MTJ consists of two ferromagnetic layers separated by an insulator sketched in Fig. 21. The TMR effect is the change of the resistance of the MTJ by switching the relative magnetization between the two ferromagnetic layers from parallel (P) to anti-

parallel (AP). The corresponding TMR ratio given by

$$TMR = \frac{g_P - g_{AP}}{\min(g_P, g_{AP})} \quad (97)$$

is a measure of the TMR effect. Thereby, g_P and g_{AP} are the conductivities of the parallel and anti-parallel magnetic alignment. The definition Eq. (97) is also called the optimistic definition because it is not restricted. Sometimes the normalized version is used, which is bounded between -1 and 1 [149, 150].

In order to use MTJs as storage elements an efficient way of switching the relative magnetic orientation is mandatory. A way with very good scalability is the STT where a current is driven through the MTJ. This current gets polarized in one magnetic layer and exerts a torque in the other magnetic layer if both magnetic layers are not perfectly aligned. This misalignment is already achieved due to temperature fluctuations. As shown in Fig. 21 there are two components of the torque, one is the in-plane and the other one is the out-of-plane component, where the plane is defined by the two magnetizations. Further, one layer is fixed in the magnetic orientation, either by thickness or by pinning. Finally, this torque can lead to switching of the so called free layer. Changing the current direction will switch back the magnetic orientation.

Now these two effects can be expanded to the field of spin caloritronics [3, 151, 152] that is by applying temperature gradients. It is clear that if a TMR effect exist there have to be also an tunneling magneto Seebeck (TMS) effect. The TMS is then the change of the Seebeck coefficient with magnetic orientation as shown in Fig. 21. Consequently, the TMS ratio is then given by

$$TMS = \frac{S_P - S_{AP}}{\min(|S_P|, |S_{AP}|)}, \quad (98)$$

where S_P and S_{AP} are the Seebeck coefficients for parallel and anti-parallel magnetic alignment. It

Spin caloric transport

30

Figure 21. Sketch of magnetic tunnel junctions (MTJs) and the principle effects. In the case of the TMR effect a bias voltage is applied and the resistance is measured for parallel (P) and anti-parallel (AP) alignment of the magnetic layers (top left). In the case of the TMS effect a temperature gradient is applied across the MTJ and a thermovoltage is measured, which gives in turn a Seebeck coefficient, which depends on the magnetic orientation of the ferromagnetic layers (top right). In the bottom the geometry for the STT is shown. One ferromagnetic layer is the fixed layer whereas the other is the free layer. The torque acting on the free layer can be decomposed into an in-plane and an out-of-plane component. The current that creates the STT can be bias voltage driven or can be a thermocurrent due to an applied temperature gradient (thermal STT).

is important to realize that the Seebeck coefficient can change sign in contrast to the conductivity. Consequently, the Seebeck coefficient can be also zero and actually does for example as a function of temperature as we will see later. In such a case the TMS ratio (98) diverges. That the TMS effect exists with a high value in a realistic MTJ was first theoretically predicted [153] and then proven experimentally [154, 155]. Since then a number of theoretical [156, 157, 158, 159] and experimental [160, 161, 162, 163, 164] works discuss the effect. Throughout this paper we will see in Sec. 7.2 that there are different strategies to optimize the effect.

One possibility which is not discussed in following is the usage of double barrier MTJs. Recently, several theoretical model calculations found interesting effects for the TMS [165, 166, 167]. However, double barrier MTJs are very hard in experiments. Also a full *ab-initio* description is cumbersome due to a lot of localized resonances.

Another effect that exploits the TMS is the spin injection into semiconductors. This effect is also called spin Seebeck tunneling [95, 168, 169]. In addition, the anisotropic TMS is discussed [170, 171]. Moreover, very recently the influence of magnons on the TMS

effect is investigated and the current interpretation is challenged [172].

Combining STT with a temperature gradient will lead to the thermal spin-transfer torque first discussed by Jia et al. [173]. The idea is quite simple. The applied temperature gradient will lead to a thermal current, which eventually exert STT on the free layer. We will discuss the effect in Sec. 7.3 and show that the effect is indeed quite small. Thus up to now a purely switching due to a temperature gradient is not realized experimentally. A number of experimental works investigate the thermal STT [174, 175, 176, 177] as well as some theoretical works [178, 179, 180, 181].

For the comparison between experiment and theory it is important to realize that in experiment a thermovoltage is measured whereas in theory the Seebeck coefficient is calculated. These two quantities are connected via the temperature drop at the junction. In the theoretical description we use linear response and thus a vanishing temperature drop. In the experiment it is hard to measure the temperature drop across the MTJ. Though first attempts were done [182, 183]. Thus, typically in experiments simulation of temperature profiles are done by using finite elements methods [154]. However, it is quite unclear if such methods can still applied at these small atomistic dimensions and how possible interface resistances are estimated. Therefore, there is a need for *ab-initio* approaches for the description of the temperature drop. Our results [184] show that the thermal conductivity of Fe/MgO is one order of magnitude smaller than expected from thin films. This finding is recently supported by experiments [185].

7.2. Tunneling Magneto Seebeck Effect

7.2.1. Basics The central quantity to calculate is the Seebeck coefficient. It is convenient to calculate it via the so called moments L_n [186]

$$L_n = \frac{2}{h} \int \Gamma(E) \left[-\frac{\partial f(E, \mu, T)}{\partial E} \right] (E - \mu)^n dE, \quad (99)$$

where $\Gamma(E)$ is transmission function and $f(E, \mu, T)$ is the occupation function. Using these moments the conductance G is simply given by

$$G = e^2 L_0 \quad (100)$$

and the Seebeck coefficient S is given by

$$S = -\frac{1}{eT} \frac{L_1}{L_0}. \quad (101)$$

Due to the two spin channels one can define a $S^\uparrow$ and $S^\downarrow$ using Eqs. (99) and (101) with the corresponding transmission functions $\Gamma^\uparrow$ and $\Gamma^\downarrow$ for both spin

channels. Within the two current model the two spin channels are conducting in parallel. Therefore their transmission function are added up in order to get the total transmission function

$$\Gamma(E) = \Gamma^\uparrow(E) + \Gamma^\downarrow(E) \quad (102)$$

Consequently, the total conductance is simply

$$G = G^\uparrow + G^\downarrow \quad (103)$$

and the Seebeck coefficient is then given by

$$S = S^\uparrow \frac{G^\uparrow}{G} + S^\downarrow \frac{G^\downarrow}{G} \quad (104)$$

Thus the total Seebeck coefficient is mainly determined by the spin channel with higher conductivity.

Looking at Eqs. (101) and (100) it becomes clear that the conductance g is directly proportional to the area under the function

$$\Gamma(E) \cdot \left(-\frac{\partial f(E)}{\partial E} \right), \quad (105)$$

whereas the Seebeck coefficient is proportional to the center of mass of the very same function. From a mathematical point of view the area under a function and the center of mass of the same function are not related. Consequently, the TMR ratio (97) and the TMS ratio (98) are independent of each other. This means that a high TMR ratio in a junction does not imply that there is also a high TMS ratio, and vice versa. In order to illustrate this fact Fig. 22 shows hypothetical transmission functions for parallel and anti-parallel magnetic alignment. Further, Eq. (105) is shown for a temperature of 300K. In this case the area under the transmission functions is quite different resulting in a large TMR ratio. At same time the centers of mass are almost identical and thus the Seebeck coefficients are the same leading to a vanishing TMS ratio. On the lower panels of Fig. 22 the TMR and TMS ratios are shown as a function of temperature, where the temperature effect is only covered by the occupation function. Clearly, the TMS ratio and TMR ratios are somehow independent. Further, it is obvious that one can construct also transmission functions where the TMR ratio vanishes but the TMS ratio is huge.

One could argue that there is the well known Mott formula [187], which connects the Seebeck coefficient to the logarithmic derivative of the conductivity. This relation can be obtained within a Sommerfeld expansion and is valid only for small temperatures or in better words if the transmission function varies only linear around the Fermi level. We will see in the next sections that this is not the case for typical magnetic tunnel junctions even at room temperatures.

Figure 22. Comparison of TMR and TMS ratio for arbitrary transmissions functions. Top left: Arbitrary transmission function for P and AP as well as the derivative of the occupation function for a temperature of 300K. Top right: The integrand (105) for P and AP for a temperature of 300K. The dots mark the center of mass. Bottom left: TMR ratio as a function of temperature. Bottom right: TMS ratio as a function of temperature.

Consequently, our argument holds that the TMR ratio and TMS ratio are basically independent of each other. Nevertheless, it might be a good starting point to use a system that already shows a large spin-dependency in transport, thus a high TMR ratio. Consequently, the starting point for theoretical and experimental investigations of the TMS were the MgO based MTJs with FeCo leads.

In any case the central quantity is the transmission function $\Gamma(E)$. Once this function is obtained, all transport coefficients (100) and (101) can be calculated. Thus all the investigations in the past targeting on tuning the transmission function in order to get a higher TMS effect. In the following sections we first explain our *ab-initio* method and then analyze the TMS in detail. After that different strategies are presented to optimize the TMS effect.

7.2.2. Methods Our *ab-initio* calculations are based on density functional theory (DFT) and we use a Green's function Koringa-Kohn-Rostoker (KKR) method. The KKR is very well suited to treat various geometries and is in particular beneficial for transport calculations [35]. In our case we use the non-equilibrium Green's function (NEGF) method in order to calculate the transmission function. Thereby, semi-infinite leads are coupled to a scattering region by self-energies of the left, Σ_L , and of the right, Σ_R lead. Eventually, the transmission function is calculated by the following trace

$$\Gamma(E) = \text{Tr} \left[\mathcal{G}(E) (\Sigma_L - \Sigma_L^\dagger) \mathcal{G}^\dagger(E) (\Sigma_R - \Sigma_R^\dagger) \right], \quad (106)$$

where $\mathcal{G}(E)$ is the retarded Green's function of the scattering region. For details of our method we refer to Refs. [188, 189, 190]. This transmission function is then used to calculate the transport properties according to Eqs. (100) and (101). Note, that the temperature dependence is covered by the occupation function only.

One big advantage of the KKR method is the description of substitutional alloys by the coherent potential approximation (CPA) [191, 192, 36, 193, 194]. Within the CPA the alloy is replaced by an effective medium, which is obtained via a self-consistent cycle. In particular, the sum of scattering properties of each individual atom within the effective medium weighted by their concentration have to be equal to the scattering properties of the effective medium. Finally, one gets an average Green's function within the effective medium.

Further, for the calculation of transport properties so-called vertex correction have to be considered [195, 196, 197, 190]. The reason is that a product of two Green's functions occurs in Eq. (106) and the average of a product is not equal two the product of the two averages. That the CPA together with vertex corrections is working have to be tested for each system. The test will be a benchmark against supercell calculations.

In our case we will apply this method to FeCo alloys. Fig. 23 clearly shows that CPA together with vertex corrections gives the same results as supercell calculations. Moreover, Fig. 23 emphasizes the great importance of the vertex corrections. Whereas for the majority spin channel the influence is rather small, it is really huge for the minority spin channel. One interpretation of the vertex correction is that it describes basically the incoherent scattering contribution. In our case this means that the conservation of the in-plane wave vector is violated due to alloy scattering. By looking at the different Fermi surfaces of the two spin channels in Fe and Co it is obvious why the minority spin is larger affected [190].

We showed the validity of the CPA including non-equilibrium vertex corrections for calculating transport properties in FeCo alloys. Thus we can apply this method as well to our MTJs. We end this section by giving one other example concerning the importance of vertex corrections. Fig. 24 shows the Seebeck coefficient of a MTJ with $\text{Fe}_{0.7}\text{Co}_{0.3}/\text{MgO}/\text{Fe}_{0.7}\text{Co}_{0.3}$ as a function of temperature with and without vertex corrections. The differences are tremendous. Thus the vertex corrections are not just small corrections. Without these corrections one obtains completely wrong results.

7.2.3. First Observations A magneto Seebeck effect was discussed in all metallic multilayer junctions by

Figure 23. Transmission at the Fermi level through $\text{Fe}_x\text{Co}_{1-x}$ as a function of x using CPA with and without non-equilibrium vertex corrections (NVC) and supercells. Taken from Ref. [190].

Figure 24. TMS for $\text{Fe}_{0.7}\text{Co}_{0.3}/6\text{MgO}/\text{Fe}_{0.7}\text{Co}_{0.3}$ as a function of temperature with (solid line) and without vertex corrections (dashed line).

Gravier et al. [198]. This effect relates to the TMS effect as the giant magneto resistance (GMR) effect relates to the TMR effect. As pointed out in Sec. 7.2.1 the TMS and TMR are not directly related but it is a good starting point to start with a material system that shows a large TMR effect.

Therefore, we calculated the TMS for Fe/MgO/Fe and Co/MgO/Co [153]. The basic finding is shown in Fig. 25. The top panel shows the Seebeck coefficient as a function of temperature for the P and AP case. For Fe/MgO/Fe both Seebeck coefficients show a change of sign with temperature. Due to the definition of the TMS ratio (98) the corresponding TMS ratio in the lower panel shows some divergences. Basically, these results show that large values of the TMS effect are possible in MgO based tunnel junctions and that there is also a large influence of the material and the temperature.

In order to understand the temperature dependence one can take a closer look at the transmission function. As an example Fig. 26 shows for the Fe

Figure 25. Seebeck coefficient for parallel S^P and anti-parallel S^{AP} alignment as well as the corresponding TMS ratios for Fe/MgO/Fe and Co/MgO/Co as a function of temperature. Partly taken from Ref. [153]

case with parallel magnetic alignment in the top panel the Seebeck coefficients of the majority and minority spin channel and in the middle panel the corresponding transmission functions. As pointed out before, in Eq. (104) the Seebeck coefficient is determined by the high conducting spin channel, which is in this example the majority spin channel. Therefore, the Seebeck coefficient of the majority spin channel is almost identical to the Seebeck coefficient of the parallel alignment. The bottom panel shows Eq. (105) for different temperatures. Again the area gives the conductivity whereas the center of mass, marked by the black dot, gives the Seebeck coefficient. Thus the competing contribution of the two peaks in the transmission function of the majority spin channel explain the observed temperature dependence.

Our prediction of the TMS effect in MgO MTJs was experimentally confirmed by Walter et al. [154] and Liebing et al. [155]. However, in both experiments a rather small TMS effect of about -9% [154] and of about 30% [155] was observed. It turned out, that the main difference between theory and between the two experiments was that in the experiments a FeCo alloy of about 50:50 [154] and of about 25:75 [155] composition was used, whereas in theory at this point only pure Fe and Co leads were considered.

Figure 26. Temperature dependent Seebeck coefficients and corresponding energy dependent transmission functions for the parallel alignment of Fe/MgO/Fe for the majority spin (left) and minority spin (right). The bottom panel illustrates the integrand (105). Taken from Ref. [153].

Figure 27. Seebeck coefficient for a random $\text{Fe}_{0.5}\text{Co}_{0.5}$ alloy at a temperature of 300K and corresponding TMS ratio as a function of the MgO barrier thickness. The dependence is given for two different temperatures.

The dependence of the composition is discussed in Sec. 7.2.6.

7.2.4. Barrier Thickness The influence of barrier thickness on the TMS ratio is not well studied up to now. For the experimentally relevant case of $\text{Fe}_{0.5}\text{Co}_{0.5}$ we show in Fig. 27 the TMS ratio as a function of the MgO thickness for two different temperatures. The TMS ratio increases first with the barrier thickness and saturates for thickness larger than 8 monolayers. This behavior is similar to the case of the TMR ratio [199].

The origin is that for thin MgO barriers the filtering effect of the barrier is not complete. Other states than the low decaying Δ_1 states contribute to transport [199]. These states will lead to a strong change of the transmission function for thinner MgO barriers. As soon as these states are damped out for a certain barrier thickness the TMS ratio will saturate.

7.2.5. Interface Termination For the TMR effect it is known that it drastically depends on the interface structure [149, 140]. Therefore, we investigated for the case of an ordered $\text{Fe}_{0.5}\text{Co}_{0.5}$ alloy different interface terminations. In particular, we considered

the structures shown in Fig. 28 with the corresponding Seebeck coefficients for P and AP alignment as well as the TMS ratios. Whereas S_{AP} is negative for all cases S_P even changes the sign with different interface termination. Additional due to the change in magnitude the TMS ratio strongly depends on the interface termination. Therefore, the interface is of special importance for the TMS effect.

The magneto-Seebeck effect in tunnel junctions with MgO barrier has also been studied for the case of both electrodes fabricated from the half-metallic Heusler alloy Co_2MnSi [201]. It is found that the magneto-Seebeck coefficient is very sensitive to the interface atomic structure. For the purely Co-terminated interface, a large effective and spin-dependent Seebeck coefficient of $-65 \mu\text{V/K}$ at room temperature is obtained from the calculations. This suggests to use such interfaces in thermally operated magnetoresistive random access memory modules, which exploit the magneto-Seebeck effect, to maximize the thermally induced readout voltage.

7.2.6. Composition Dependence In Sec. 7.2.3 we discussed that there is a large difference between the TMS ratios obtained in experiment and in theory. Thereby, the main difference is that in our calculation we used only pure Fe or Co or a perfectly ordered $\text{Fe}_{0.5}\text{Co}_{0.5}$ alloy. In order to overcome this drawback we employ the CPA together with vertex corrections to calculate the TMS ratio for different random FeCo alloys [202]. Fig. 29 top panel shows the Seebeck coefficients for the P and AP case for two different MgO barriers. The influence of the barrier thickness is rather small except close to the pure materials. The dependence of the Seebeck coefficient for the P case can be clearly attributed to a shift of the Fermi level due to the alloying [202]. For the AP case the situation is more complex.

For both magnetic alignments the Seebeck coefficient shows a number of sign changes, which lead to a strong dependence of the TMS ratio on the composition, which is shown in the bottom panel of Fig. 29. These sign changes lead to divergencies of the TMR ratio. The important results from Fig. 29 is that at composition at around 50:50 FeCo the TMS ratio is rather small, only a few percent. This is the typical composition in experiments. Therefore, the obtained experimental results can be explained by the calculations. Further, the TMS ratio can be tuned by the composition.

We will end this section by giving a comparison to the composition dependence of the TMR ratio [156]. This is shown in Fig. 30. Thereby, the TMR ratio is rather constant as a function of the alloy composition. This means that the TMS ratio can be

Figure 28. Seebeck coefficients and corresponding TMS ratios as a function of temperature for different interface terminations (partly taken from Refs. [200] and [154]).

Figure 29. Seebeck coefficients and corresponding TMS ratio as a function of $\text{Fe}_x\text{Co}_{1-x}$ composition for two different MgO thicknesses. The dashed lines for the TMS ratio are obtained by using Eq. (98) with linearly interpolated values of the Seebeck coefficients. Taken from Ref. [202]

Figure 30. TMR ratio as a function $\text{Fe}_{1-x}\text{Co}_x$ composition for zero and a finite bias voltage. Taken from Ref. [156]

more sensitive to changes in material than the TMR ratio. Consequently, for certain applications, e.g. as a sensor, the TMS effect might be beneficial.

7.3. Thermal Spin Transfer Torque

7.3.1. Basics and methods The idea of the thermal STT is the usage of a temperature gradient instead of an applied bias voltage to switch the free layer. This principle was already sketched in Fig. 21. The temperature gradient creates a thermocurrent which finally exerts a STT on the free layer. Other possibilities such as the use of magnons [6] are not discussed here. The torque τ_i acting on layer i at a given energy E is

given by [203, 188]

$$\tau_i(E) = \frac{1}{\hbar} \Delta_i \times \delta \mathbf{m}_i(E) , \quad (107)$$

where Δ_i and $\delta \mathbf{m}_i(E)$ are the exchange field and the magnetization of the transport electron in layer i , respectively. As in the previous section we use our KKR method together with the CPA method including vertex correction to calculate the magnetization of the transport electron. Using the NEGF method the magnetization of the transport electron can be calculated. This can be used in Eq. (107) to calculate the torque. Details about our method can be found in Refs. [188, 190].

Now the torque can be decomposed in an in-plane and out-of-plane component. The plane is defined by the two magnetizations. In the following we want to focus solely on the in-plane component, because it is the larger component and more important for switching [148]. Note that we omit for clarity any additional index, but in the following we only talking about the in-plane component.

In order to get the total torque τ_{tot} acting on the free layer we have to integrate the torque originating from electrons coming from the left $\tau_{L \rightarrow R}$ and the torque originating from electrons coming from the right $\tau_{R \rightarrow L}$ over energy

$$\tau_{tot} = \int [\tau_{L \rightarrow R}(E) f_L(E, \mu_L, T_L) + \tau_{R \rightarrow L}(E) f_R(E, \mu_R, T_R)] dE , \quad (108)$$

where $f_{L(R)}(E, \mu_{L(R)}, T_{L(R)})$ is the occupation function of the left (right) lead, which is at temperature $T_{L(R)}$ and electrochemical potential $\mu_{L(R)}$. In the case of ballistic and elastic transport the torque fulfills

$$\tau_{L \rightarrow R}(E) = -\tau_{R \rightarrow L}(E) . \quad (109)$$

Thus Eq. (108) simplifies to

$$\tau_{tot} = \int \tau_{L \rightarrow R}(E) [f_L(E, \mu_L, T_L) - f_R(E, \mu_R, T_R)] dE . \quad (110)$$

This equation is valid for applying a bias voltage as well as for applying a temperature gradient. The main differences between both cases are the difference of the occupation functions. To illustrate this fact Fig. 31 shows the difference of the occupation functions for the case of a temperature gradient and for the case of an applied bias voltage.

It is obvious that for the case of the applied bias voltage we basically get the area under the energy dependent torque whereas for the temperature gradient it is basically the asymmetry of the energy dependent torque. In the typical case where one spin channel is dominant the torque has the same sign around the Fermi level, which means that the bias voltage driven STT is larger than the thermal STT.

Figure 31. Difference of the occupation functions used in Eq. (110) for the case of an applied bias voltage of 68mV (red) and for the case of an applied temperature gradient of 100K (black). For the latter the temperature of the left and right lead are 200K and 100K, respectively. Taken from Ref. [204].

Figure 32. Angular dependence of the thermal STT. Taken from Ref. [204].

7.3.2. Results Due to the small expected thermal STT it is necessary to use very thin MgO barriers. Thereby, a critical thickness in experiments is 3 monolayers of MgO [174]. For such thin barriers Jia et al. [173] showed that the angular dependence deviate from a sine dependence. The origin is that for thin MgO barriers the spin filtering of the Fe/MgO is not as effective as for thicker barriers. Consequently, more states contribute within the Brillouin zone and thus a more complicated angular dependence arises. This is confirmed by our calculations shown in Fig. 32.

In a next step we analyze the dependence of the thermal STT for different FeCo compositions. Fig. 33 shows for a fixed temperature of the left lead the dependence of the torque on the temperature gradient. Obviously, the composition has a huge impact on the size of the thermal STT. It even changes the sign of

Figure 33. Thermal STT at an angle of 90° of the magnetizations as a function of temperature gradient for different $\text{Fe}_x\text{Co}_{1-x}$ compositions. The temperature of the left lead is fixed at $T_L = 300\text{K}$. Taken from Ref. [204].

the slope.

Similar to the comparison of TMR and TMS the bias voltage driven STT is basically independent of the composition [156]. Thus the thermal STT is way more sensitive to material changes, because it is a measure of the asymmetry. This also means that there might be a large playground to optimize the effect further.

In summary, the thermal STT is a very small effect. Consequently, very thin barriers and very thin free layers are needed. But even then very high temperature gradients are needed in the order of several Kelvin [173]. All these requirements are basically independently possible in experiments, but up to now not in combination [174].

7.4. Conclusions

Many studies were carried out to investigate the TMS, but a number of questions still need to be addressed in the future, in particular by *ab-initio* theory. One very important aspect is the role of magnons, which is basically not clear. From models [172] there might be some important influence but only *ab-initio* theory can clarify this point.

Further, the TMS can be further optimized. However, for an optimization one basically need some kind of application first in order to define an optimization goal. Besides a high TMS ratio, maybe high output voltages or high sensitivity to applied bias voltages or to structural parameters are needed. In this respect, our results shows that the TMS effect is more sensitive than the TMR effect and thus might have some advantages for certain applications.

More fundamental the temperature profiles at the nanoscale are a complex issue on its own. Here efforts on theoretical as well as experimental side

are necessary for a real understanding of temperature profiles at the nanoscale.

8. Effect of spin disorder at elevated temperatures on the transport properties of magnetic nanostructures

Natural candidates for the materials design with tailored spin-caloric properties are the magnetic nanostructures with the spin relaxation length possibly longer than the nanostructure diameter (yielding less volatile spin transfer) and pronounced quantum confinement effects suggesting that extraordinary spin-caloritronic effects could be realized [205, 206, 207, 208, 209, 210, 211, 212]. The electron-transport phenomena in magnetic materials at high temperatures are, however, strongly affected by the formation of a spin-disordered state as a result of local-moment fluctuations which induce spin-conserving and spin-flip scattering. The well-known *spin disorder* resistivity in ferromagnetic materials has been experimentally studied in the past [213, 214] and *ab-initio* techniques were successfully used to model this effect [215, 216, 217, 218, 219]. The moment fluctuations in bulk are small for temperatures significantly lower than the critical temperature, but the surface-to-volume ratio in nanostructures is large and the fluctuations are correspondingly stronger. The spin disorder obviously contributes also to thermoelectric and spin-caloric phenomena, yet its influence in the current spin polarization and in the Seebeck and spin Seebeck effects was not previously investigated. In the following, we summarize the most important findings of the studies addressing this issue by means of the combination of density-functional calculations and Monte-Carlo simulations [220, 221].

8.1. Electron transport with real space spin disorder

The magnetic nanostructures are modeled based on the following geometry. The nanostructures are embedded between free-electron-metal leads and either forming a two-dimensional thin layer or being surrounded in the lateral plane by a free-electron metal, vacuum or an insulating material. The general setup is shown in Fig. 34. The fcc crystal lattice (typically Cu or Ag atoms) forms the left and right (or top and bottom) semi-infinite leads interfacing the scattering region in which the fcc lattice is followed epitaxially, which is justified by its small extent. We do not perform atomic relaxations as our objective is to study the effect of spin disorder.

A supercell in the xy plane is employed to model the real space spin disorder in thin layers and also in nanowires, separating them adequately from their periodic images [220, 221]. The transmission

Figure 34. (Color online) General setup of the model systems with the direction of current parallel with the z axis. The sites where the temperature induced spin disorder is considered are shown as arrows. The transmission probability is evaluated for pairs of atomic layers (indicated by vertical lines) placed far enough from the interface between the spin ordered lead and spin disordered scattering regions. Adapted from Fig. 1 of Ref. [220].

probability between the two leads is calculated for pairs of atoms in atomic layers which are chosen at a distance enough far away from the interface with the scattering region as to ensure their bulk-like potentials [222]. The disorder effect in diverse material properties was previously modeled employing the supercell approach, e.g., the spin-disorder resistivity [215, 217, 218, 219], or the frozen thermal lattice disorder effect on the Gilbert damping and the spin-flip diffusion length [223, 224]. The disordered local moment method in the coherent potential approximation, begin computationally cheap, can be also used to evaluate the spin disorder effect but a complete disorder saturation above the critical temperature due to the missing short range order can lead to significant differences in the results [216].

Electronic structure calculations were performed by using the Korringa-Kohn-Rostoker Green-function method (KKR-GF) employing the local density approximation [225] to the exchange-correlation energy functional and the full-potential formalism [226, 227, 228, 229] with the truncation of the angular momentum expansion at $l_{\max} = 3$. The spin-orbit coupling is not considered as already at low temperatures the thermally induced spin fluctuations become a dominant effect.

We model the spin disorder by applying a classical Heisenberg model

$$H = - \sum_{i,j} J_{ij} \mathbf{M}_i \cdot \mathbf{M}_j, \quad (111)$$

where J_{ij} are the exchange parameters extracted from the ground-state electronic structure following the method of infinitesimal rotations [230] and $\mathbf{M}_i$ and $\mathbf{M}_j$ are unit vectors pointing in the direction of the magnetic moments at temperature T at sites i and j , respectively. The moment directions are sampled in a series of “snapshots” at thermal equilibrium at T by means of the Monte Carlo (MC) method using the Metropolis algorithm [231] and the Mersenne twister [232] for the random number generation. The

number of required MC configurations is proportional to the magnetization fluctuation amplitude. Besides the magnetic susceptibility, the moment-moment correlation function C_N between moments, averaged over all layer pairs having a distance of N layers [220, 221], is used to characterize the loss of magnetic order.

The calculation of transmission probability makes use of the adiabatic approximation [233, 234], with the assumption that the energy is not exchanged between electrons and the magnetic system while the electrons traverse the nanostructure. Furthermore, during this fast process the magnitude and direction of the magnetic moments are treated as frozen. This was successfully applied in the past for the spin-disorder resistivity calculation in ferromagnets in e.g., Refs. [217] and [218]. The computer code to evaluate the transmission probability matrix through the non-collinear magnetic structure [220, 221] follows a combination [222] of the KKR-GF method [226, 35], and the Baranger-Stone [130] Green function approach to the Landauer-Büttiker theory.

A non-self consistent rotation of the ground-state magnetic part of the site-dependent potentials further approximates the magnetic moment instantaneous direction prescribed by the MC. This was thoroughly tested and only negligible differences were found between the magnitude of the magnetic moments in the non-collinear state and the ground state, indicating the dominance of the intra-atomic exchange interaction over the inter-atomic interaction for the moment formation and justifying the non-self-consistent approximation. For brevity, the majority and minority spin channel will be referred to as $\uparrow$ and $\downarrow$, respectively, and taken with respect to the global reference frame defined by the effective moment direction of the model system [220, 221].

Following the idea of multiple-scattering theory and the KKR-GF method, the site-dependent t -matrices are rotated in spin space and the Dyson equation is used to obtain the retarded Green function $\mathcal{G}(E)$ for the magnetically non-collinear system. Considering the periodic supercell geometry, the Fourier transformation yields the Green function for each momentum channel $\mathbf{k}_{\parallel}$. The resulting transmission probability matrix in spin space as a function of $\mathbf{k}_{\parallel}$ and E is evaluated as

$$\begin{aligned} \Gamma^{\sigma\sigma'}(\mathbf{k}_{\parallel}, E) = & \sum_{\mu\mu'} \sum_{LL'L''L'''} (J_{LL'}^{\mu\sigma} - J_{L''L'''}^{\mu\sigma*}) \\ & \times (J_{L'L''}^{\mu'\sigma'} - J_{L''L'''}^{\mu'\sigma'*}) \\ & \times \mathcal{G}_{LL'}^{\mu\mu'\sigma\sigma'} \mathcal{G}_{L''L'''}^{\mu\mu'\sigma\sigma'*}, \end{aligned} \quad (112)$$

where the current-density matrix elements $J_{LL'}^{\mu\sigma}$ in the non-magnetic lead are associated with the cell at site μ , angular momentum indices L , and spin indices σ . Atomic pairs of the cells in the left and right lead

are chosen to cover the whole cross-section of the leads (see Fig. 34). Well converged results in the fcc lattice system are achieved already by using a single atomic layer on each side [222]. The artifacts of the calculations of $\Gamma^{\sigma\sigma'}(\mathbf{k}_{\parallel}, E)$ due to the complex energy contour are removed by an extrapolation of the values to the real energy axis E [220, 221].

The transport coefficients $\mathbf{L}_n$ as 2×2 matrices in spin space are evaluated by a numerical integration of $\Gamma(\mathbf{k}_{\parallel}, E)$ over the crystal momentum $\mathbf{k}_{\parallel}$ and energy E as

$$\mathbf{L}_n = \int dE \Gamma(E) \left[-\frac{\partial f^0(E, \mu, T)}{\partial E} \right] (E - E_F)^n \quad (113)$$

where $f^0(E, \mu, T)$ is the Fermi-Dirac distribution, T is the temperature of the MC simulation, and $\mu = E_F$ is the Fermi energy. For each temperature and system, $\Gamma(E) = \int_{\text{SBZ}} d\mathbf{k}_{\parallel} \Gamma(\mathbf{k}_{\parallel}, E)$ was calculated on a mesh of $2N_E + 1$ equidistant energy points ($N_E \approx 7 - 10$) in the range $-N_E k_B T \leq (E - E_F) \leq +N_E k_B T$, beyond which $(E - E_F)^n \partial f^0 / \partial E$ practically vanishes. Tests on denser grids gave insignificant differences. Finally, $\langle \mathbf{L}_n \rangle_T$ is calculated as an average over the non-collinear MC configurations. The electrical conductance G , electrical resistance R , charge Seebeck coefficient S and spin Seebeck coefficient S^{spin} , the thermal conductance K , and the thermoelectric figure of merit ZT are calculated as

$$G^{\sigma\sigma'}(T) = \frac{e^2}{h} \langle L_0^{\sigma\sigma'} \rangle_T ; \quad G = \sum_{\sigma\sigma'} G^{\sigma\sigma'} ; \quad R = \frac{1}{G} \quad (114)$$

$$S = -\frac{1}{eT} \frac{\sum_{\sigma\sigma'} \langle L_1^{\sigma\sigma'} \rangle_T}{\sum_{\sigma\sigma'} \langle L_0^{\sigma\sigma'} \rangle_T} \quad (115)$$

$$S^{\text{spin}} = -\frac{1}{eT} \frac{\langle L_1^{\uparrow\uparrow} \rangle_T + \langle L_1^{\downarrow\downarrow} \rangle_T - \langle L_1^{\uparrow\downarrow} \rangle_T - \langle L_1^{\downarrow\uparrow} \rangle_T}{\sum_{\sigma\sigma'} \langle L_0^{\sigma\sigma'} \rangle_T} \quad (116)$$

$$K = \frac{1}{T} \left[\sum_{\sigma\sigma'} \langle L_2^{\sigma\sigma'} \rangle_T - \frac{\left(\sum_{\sigma\sigma'} \langle L_1^{\sigma\sigma'} \rangle_T \right)^2}{\sum_{\sigma\sigma'} \langle L_0^{\sigma\sigma'} \rangle_T} \right] \quad (117)$$

$$ZT = \frac{GT}{K} S^2. \quad (118)$$

8.2. Co nanostructures between Cu leads

The magnetic nanostructures consist of 8 atomic layers of either Co atoms forming a thin layer or Co atoms in a shape of a thin wire surrounded by Cu or vacuum (see Fig. 35). The leads are made of fcc lattice of Cu atoms with the experimental lattice constant $a_{\text{lat}} = 3.62 \text{ \AA}$. Here, we will present only results for the thin layer (TL) and diatomic wires (W22), the influence of other structural configurations are discussed in Ref. [220].

Figure 35. (Color online) Cobalt (blue/dark spheres) nanostructures sandwiched between Cu leads (yellow and red/bright spheres): (a) thin Co layer “TL” consisting of 8 monolayers, (b,c) diatomic wires “W22(Cu)” and “W22(Va)” having 2 Co atoms in all layers. The (Cu) and (Va) indicate the type of embedding: copper and vacuum, respectively. Periodic boundaries are indicated by solid lines. Crystal structures were plotted using VESTA [235]. (d) Setup of the W22 nanowire geometry: Co sites (red / filled circle), their first nearest neighbor (green / thick line circle), second nearest neighbor (blue / medium thick line circle) and embedding sites (black / thin line circle) are shown in the two adjacent layers (large and small circles, respectively). The 3×3 and 4×4 supercell is indicated by solid and dashed line, respectively. Adapted from Fig. 2 of Ref. [220].

The ground state electronic structure is determined in a three-step procedure. In the first step, the density and potential of a $\text{Cu}(7)\text{-}\{\text{Cu/Co/Va}\}(8)\text{-Cu}(7)$ slab with one atom per layer is calculated self-consistently. Depending on the model system, the scattering region $\{\text{Cu/Co/Va}\}(8)$ contains eight layers of Co atoms, Cu atoms or vacuum. The scattering region is sandwiched between two leads with seven atomic layers of Cu. The slab is embedded in three layers of empty spheres on each side of the slab, modelling vacuum. In the second step, the outermost part of the slab (three outermost Cu atomic layers and the outer vacuum) is replaced by half-infinite Cu leads using the decimation technique [236, 237] in the KKR-GF method [238, 226]. In the final step, the supercell potentials are constructed. A replication of the individual site potentials within the 3×3 supercell [Fig. 35(a)] yields the TL system. The W22 nanowires were likewise modeled in the 3×3 supercell [Fig. 35(b,c,d)]. To obtain their electronic structure, the impurity Green function method [239, 226] was used to determine the self-consistent potentials in the Co wire cells and their nearest neighbors. More detailed description of the procedure can be found in Ref. [220].

The exchange coupling parameters J_{ij} were

evaluated between the Co atoms with the maximum distance of $3 a_{\text{lat}}$. No effect was found by including the J_{ij} parameters with larger interatomic distances. As no real critical temperature can be determined in the nanostructures, a crossover temperature at the peak of the magnetic susceptibility χ is used to define T_c . In the TL system, the critical-like character is found at $T_c \approx 1100$ K. In contrast, a rather large magnetic moment is preserved in all nanowires until very high temperatures, resembling macro-spin character and the divergence of χ is only weak with its maximum shifted to much lower temperatures. The spatial correlation $C_N(T)$ was generally found to be a much better indication of the long-range magnetic order loss. The crossover point in the TL system, with the most bulk-like character, was calibrated by $C_3 < 0.12$ condition [dashed vertical line in Fig. 36(a)]. In the nanowire systems, the difference between the T_c and a crossover point determined from the C_3 falloff is rather large indicating a more gradual long-range order loss.

In Figure 36(a), we present the well-known effect of spin disorder resistivity. A clear correlation between the characteristic kink in the electrical resistance of the TL and its crossover point can be seen. In the W22 embedded in Cu, the not so sharp kink is also present and the resistance ultimately saturates. Even less pronounced kink is found in the W22 embedded in vacuum and the resistance keeps growing above the crossover temperature. The polarization of the electrical conductance [Fig. 36(b)] shows that the nanowire embedding can have a very large effect. Moreover, the spin disorder indeed shows a significant effect (solid line), e.g., the strong suppression of the polarization or the sign reversal in W22(Va).

While the character of electrical conductance can be, for some simple systems, following the character of their electronic density of states (DOS), it is virtually impossible to recover such connection for the Seebeck coefficient (except maybe in the low-temperature limit in some model systems). The simple argument behind this observation is that the Seebeck coefficient is a ratio of the two integral quantities, the L_1 and L_0 transport coefficients, where the latter could be already unrelated to the DOS and the former is composed of contributions of the transmission probability with maximum weight at $|E - E_F| \approx 1.5 k_B T$ and significant weight up to as far as $|E - E_F| \approx 5 k_B T$.

The conventional charge (S) and spin (S^{spin}) Seebeck coefficient is shown in Fig. 36(c) and Fig. 36(d), respectively. The temperature influence is generally non-trivial, even in the spin ordered case (dotted line). The Seebeck coefficient is negative in TL and W22(Cu) and its sign is reversed in W22(Va). This observation can be easily traced back to the asymmetry

Figure 36. (Color online) (a) Total electrical resistance. Vertical lines indicate temperature for which $C_3 < 0.12$. (b) Polarization of the electrical conductance. (c) Charge and (d) spin Seebeck coefficient. Dotted and solid lines correspond to the spin-ordered and -disordered data, respectively. The color coding of data sets in the individual columns corresponds to the system labels in Fig. 35. Adapted from Fig. 5 of Ref. [220].

of $\Gamma(E)$ around E_F . In the case of TL and W22(Cu), the $L_1^{\uparrow\uparrow}$ and $L_1^{\downarrow\downarrow}$ have an equal sign, in W22(Va), the sign of $L_1^{\uparrow\uparrow}$ and $L_1^{\downarrow\downarrow}$ is opposite and the $L_1^{\downarrow\downarrow}$ grows faster with temperature than $L_1^{\uparrow\uparrow}$.

Quantum confinement was found as the Van Hove feature in the DOS of the nanowires, but it was not reflected in the transport properties. In Ref. [240], the minority-spin channel quantum well states of the Cu/Co interface were found to affect the transport properties as a function of the Co layer thickness in the ideal spin ordered case. If spin-disorder is present leading to strong mixing of the two spin channels, such effects of the quantum confinement should be reconsidered in detail.

The spin disorder effect on the Seebeck coefficients [solid line in Figs. 36(c,d)] is generally rather significant, similarly to the electrical resistance and the conductance polarization. The strong enhancement of S in W22(Cu) at higher temperatures is a direct consequence of the $\Gamma(E)$ modulation, where a kink at $\Gamma^{\uparrow\uparrow}$ located near the E_F increases the $L_1^{\uparrow\uparrow}$ asymmetry. As anticipated, the spin disorder leads to a strong suppression of the spin Seebeck coefficient, although the S^{spin} remains relatively large at high temperatures in W22(Cu) and W22(Va) nanowires as a result of the non-vanishing difference between the $\uparrow$ and $\downarrow$ L_1

Figure 37. (Color online) (a) Ag/(Cr,Zn)Te interface unit cell, the layers are ordered along the z direction as Te, Cr/Zn and Ag. The CrTe nanostructures sandwiched between Ag leads (oriented according to the xyz compass): (b-d) CrTe thin layers (TL) of a variable thickness and monoatomic Cr nanowires (W1) embedded in ZnTe matrix with a substitutional impurity in its middle (e). Different elements in (b)-(e) are depicted as large dark spheres (Cr), large bright spheres (Zn), medium size spheres (Te) and small spheres (Ag). The number of Cr layers is 13 (b) and (e), 9 (c), and 5 (d). The structure representations were plotted using VESTA [235]. Adapted from Fig. 1 of Ref. [221].

transport coefficients.

8.3. CrTe nanostructures between Ag leads

The magnetic nanostructures consist of the scattering region with the zinc blende (ZB) crystal structure (experimental lattice parameter of ZB-ZnTe $a_{\text{lat}} = 6.1 \text{ \AA}$) which are sandwiched between Ag leads (Fig. 37). The scattering region is filled either by a thin layer (TL) of CrTe or by a monoatomic wire (W1) of CrTe laterally embedded in the ZnTe matrix. The nanostructures are terminated with Zn/Cr layer interfacing the leads. The interface layer Ag atoms continue the ZB structure at the positions of Te and vacant sites as can be seen in Fig. 37(a), depicting the interface in-plane unit cell with lattice constant of $a_{\text{lat}}/\sqrt{2}$. A 3×3 in-plane supercell was employed for the real space disorder model in the CrTe TL as well as a separation between the W1 periodic images. The supercell potentials were constructed following the procedure described in Section 8.2. The effect of the TL thickness was examined in the systems with 13, 9 and 5 Cr layers, indicated by a number in parentheses, e.g. TL(9). More details and the effect of the W1 length (not discussed here) can be found in Ref. [221].

The collinear magnetic ground state was used in the self-consistent calculations of the potential,

however, an analysis of the exchange coupling parameters (not shown here) gave us an indication towards a tendency to form a non-collinear ground state in the Cr interface layer. This was indeed confirmed in the MC simulations. The directions of Cr magnetic moments at the interface tilt away from the film-interior magnetization, forming a checkerboard pattern, but do not reach a fully spin-flop state. This non-collinearity is accounted for in the calculations at finite temperatures.

In Figure 38(a), the characteristic kink in the $R(T)$ due to the loss of the long range order can be identified and associated with the $C_3 < 0.1$ condition at 400 K in the most bulk-like TL(13) system. A slow $C_2(T)$ falloff is consistent with a further milder increase of the resistance at higher temperatures until the resistance saturates. We estimate the spin disorder contribution to the resistivity from the slope of the resistance as a function of the TL thickness (lead-to-lead distance) at $T = 500 \text{ K}$ with almost fully saturated spin disorder. Assuming that the Ohmic limit is fulfilled for the thickness of 9 Cr layers [217, 218], the spin disorder contribution to the resistivity $\rho_{\text{sd}} = N_{\text{Cr}} a_{\text{lat}} (R_{500 \text{ K}}^{\text{TL}(13)} - R_{500 \text{ K}}^{\text{TL}(9)})/4$, where $N_{\text{Cr}} = 9$ is the number of Cr atoms in the supercell cross-section. We obtain a value of $12 \mu\Omega \text{ m}$ in a reasonable agreement with the experimentally obtained estimate of the joint spin disorder and phonon contribution to the resistivity ($20 \mu\Omega \text{ m}$) in bulk $\text{Cr}_{0.9}\text{Te}$ (NiAs structure) 400 K [241] and also with the more recent resistivity measurement of the CrTe thin film at room temperature ($\approx 10 \mu\Omega \text{ m}$) [242].

Considering the non-collinear state at the interface at $T \rightarrow 0$, while the half-metallicity of the CrTe film interior is still preserved, a finite probability exists for a spin-down electron to traverse between the leads due to the spin-flip processes occurring at the interface. This effect yields an increase in the conductance (decreased resistance) at low T in comparison with the collinear case as shown in Fig. 38(a), particularly apparent in the thin TL(5) system. While in thicker systems this effect can be strongly masked when only the resistance is analyzed, an almost identical signature is recognized in the conductance polarization, irrespective of the TL thickness [Fig. 38(b)]. The increasing temperature yields a very similar effect in the conductance polarization of the TL systems and becomes negligible above a kink at the cross-over temperature.

The charge-Seebeck coefficient S of the TL systems [Fig. 38(c)] in the collinear case is very low as a result of the low asymmetry in $\Gamma(E)$. The spin disorder affects all TL systems in a similar way, S becomes consistently positive and keeps growing with the temperature. The spin-Seebeck coefficient S^{spin}

Figure 38. (Color online) (a) Electrical resistance multiplied by the number of Cr atoms in the supercell cross-section ($N_{Cr} = 9$) for TL systems. Vertical lines indicate the temperature for which $C_3 < 0.1$. (b) Polarization of the electrical conductance. (c) Charge- and (d) spin-Seebeck coefficient. Dotted and solid line corresponds to the spin-ordered and spin-disordered data, respectively. Adapted from Fig. 3 of Ref. [221].

[Fig. 38(d)] follows the trends in the S resulting from the spin disorder, except that it is suppressed with the increasing temperature and becomes already very low at the room temperature.

An investigation of the substitutional impurity effect as a source of extra scattering in the monoatomic CrTe nanowires was motivated by the strong $\Gamma(E)$ modulation around the E_F . The W1(13) nanowire was selected as a representative due to the robustness of its Seebeck coefficients with respect to the spin disorder (see Ref. [221] for more details). An asymmetry enhancement of the $\Gamma(E)$ peak around E_F can be expected resulting from an energy shift or a shape modulation under the impurity presence. The Cr atom in the W1(13) center was systematically substituted by elements of the fourth period, from potassium to germanium [Fig. 39]. The electronic structure was determined in a self-consistent way by means of the impurity Green function method [239, 226].

Taking the already mentioned robustness of the W1(13) Seebeck coefficient against the spin disorder into consideration, we first examined the transport properties in the spin ordered case where the temperature of 470 K was accounted for by only the Fermi smearing. For all elements from Ti to Ni, we found a sizable magnetic moment at the impurity site [Fig. 39(a)]. The magnetic moments of Ti and

Figure 39. (Color online) Properties of the W1(13) nanowire with the substitutional impurity in its central layer. (a) Majority spin d orbitals occupation of the impurity site (solid line). Magnetic moment $m(\mu_B)$ of the impurity site (if $m > 0.4 \mu_B$) is represented by a vertical arrow, pointing up/down in case of parallel/antiparallel orientation with the nanowire magnetization. (b) The $k_{\parallel}$ -integrated transmission probability as a function of the energy around E_F (vertical dashed line). The values of $E_F \pm 3k_B T$ are indicated by the ticks on the E axis. (c) The transport coefficients L_0 and L_1 ($R_K = h/e^2$ is the von Klitzing constant). (d) Thermoelectric figure of merit. Adapted from Fig. 6 of Ref. [221].

Table 3. Charge and spin Seebeck coefficients (in $\mu V/K$) and figure of merit for the W1(13) nanowire with selected substitutional impurities (Sc and Ti). Note that in the spin ordered case, $S \approx S^{spin}$ due to negligible $L_n^{\downarrow\downarrow}$, $L_n^{\uparrow\downarrow}$ and $L_n^{\uparrow\uparrow}$ terms in comparison to $L_n^{\uparrow\uparrow}$ for both $n = 0$ and $n = 1$. Taken from Ref. [221].

impurity	spin order		spin disorder		
	$S \approx S^{spin}$	ZT	S	S^{spin}	ZT
Sc (290 K)	71	0.27	80	38	0.35
Sc (470 K)	56	0.21	58	25	0.19
Ti (290 K)	47	0.09	58	26	0.12
Ti (470 K)	59	0.18	64	28	0.18

V impurities were aligned in parallel with the overall nanowire magnetization, whereas for the Mn, Fe, Co and Ni impurities the anti-parallel alignment was energetically preferred. As a result, the majority spin d character occupation of the Mn–Ni sequence is qualitatively similar to the Sc–Cr sequence. A significant asymmetry of the $\mathbf{k}_{\parallel}$ -integrated Γ around E_F can be indeed identified for certain impurities in Fig. 39(b). The strong asymmetry is manifested in the L_1 transport coefficient in case of Sc, Ti and Ni impurities while the L_0 coefficient is generally decreased by the presence of all impurities except Ni [Fig. 39(c)]. The S enhancement by an order of magnitude in comparison with the pure W1(13) nanowire is found in case of the Sc and Ti impurities with the corresponding two orders of magnitude increase of ZT , reaching values of about 0.2 [see Fig. 39(d) and Table 3]. For the Sc and Ti impurities, the influence of spin disorder was further examined at the room (290 K) and elevated (470 K) temperatures. The $\Gamma(E)$ asymmetry was affected only weakly but the slightly reduced conductance yields a further enhancement of the S with the largest ZT of 0.35 observed for the Sc impurity at the room temperature. The spin disorder brings a reduction of about 45% to the S^{spin} with respect to the S but it keeps sizable values at elevated temperatures.

8.4. Summary and conclusions

We investigated the transport properties considering the effect of temperature induced spin-disorder in several magnetic Co nanostructures embedded between Cu leads and CrTe nanostructures embedded between Ag leads. It was found that the spin disorder affects the transport coefficients both qualitatively and quantitatively at elevated temperatures, and therefore cannot be neglected in a theoretical analysis. Simple models of the electronic temperature, such as the Fermi smearing, does not lead, in many cases, to even qualitatively adequate description of the transport coefficients. Unfortunately, the validity of this simplified and computationally less expensive approach cannot be evaluated beforehand; the verification comes only *a posteriori*, in comparison with the calculation where the spin disorder was explicitly included.

Calculations of the charge and spin Seebeck coefficient as a function of temperature exhibit a non-trivial behavior due to a number of effects that are factored in: the local magnetic moments fluctuations, their temperature-dependent correlation, the nanostructure geometry leading to a possible quantum confinement effects, the conducting states entering via the Fermi distribution, and the interface transmission. We also showed that the charge and spin Seebeck coefficients can be increased, decreased or even

reverse a sign as a result of an interplay between the particular microscopic structure and the spin disorder. The thermoelectric figure of merit ZT can be enhanced by orders of magnitude either by the spin disorder effect or by a substitutional impurity of a particular element in the magnetic nanostructure. Additionally, no clear connection between the density of states and the transport properties (resistance, charge and spin Seebeck coefficients) is found, due to the convolution of the energy dependent transmission probability and the derivative of the Fermi function.

Finally, our CrTe thin-film calculations address the issue of the usefulness of half-metallic ferromagnets in order to produce spin polarized electron transport. We found that if a non-collinear magnetic phase develops at the interface of a half-metallic magnet to the leads, the current spin polarization drastically drops. Additionally, the current spin polarization is further reduced by the non-collinear state at $T > 0$ due to local-moment fluctuations.

9. Conclusions and outlook

A plethora of different problems in spin caloric transport are accessible by DFT based methods. In effect, these include all phenomena that derive from electronic transport. The Seebeck, magneto-Seebeck and tunneling magneto-Seebeck effects, the spin-Nernst effect, the thermal spin transfer and thermal spin-orbit torque, all these phenomena are described while accounting for the complexity of the electronic structure without adjustable parameters. The effects of interface scattering, chemical disorder, spin disorder and atomic vibrations are within the reach of the DFT methods and may be switched on or off at will in numerical experiments, revealing the relative importance of the various contributions. Corrections are needed in the cases where the approximations to exchange and correlation are poor within the DFT description of the ground state, for example in strongly correlated systems.

A direct comparison with experiment is not always straightforward, and requires targeted design of experiments, under controlled conditions required as input by *ab-initio* theory (with respect to sample composition and disorder, interface structure, temperature gradients, etc.) Contrary to model-based or *ad hoc* approaches, that depend on adjustable parameters, *ab-initio* calculations are, in effect, numerical experiments that necessitate a thorough description of the experimental setup on a microscopic level. The experimental setup is frequently complicated, for instance by the indirect read-out of the spin current by means of the inverse spin-Hall effect, or not fully controlled, for instance due to

unknown temperature profiles or unknown interface roughness in junctions. Possible targeted experimental design would include measuring the trends in alloys with respect to composition or concentration. Still, *ab-initio* theory and experiment have worked hand-in-hand in order to clarify questions on the microscopic origin of spin caloric effects, e.g. in tunnel junctions [154, 202] or in ferromagnetic metals [85].

Concerning thermal and spin transport by means of phonons and magnons, that was not reviewed here, we should expect major effects in magnetic insulators, as well as in metal-insulator interfaces by electron-phonon scattering. The latter can give corrections in the case of tunnel junctions. DFT methods can contribute to the understanding of these phenomena by employing *ab-initio* molecular dynamics and spin dynamics methods, which, however, is a numerically very expensive task. More realistic is to employ dynamical model calculations [243], where the model parameters (force constants and phonon dispersion [244], or exchange interactions between magnetic moments and magnon dispersion [230, 233, 234]) are calculated within the DFT.

A combination of the above complementary methods should describe the full spectrum of effects in spin caloric transport and contribute, on the one hand, to the prediction of new materials or new functionalities, and on the other hand, to the complete, quantitative interpretation of experiments.

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