Topical Review

Tunnel magneto-Seebeck effect

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Abstract
The interplay of charge, spin and heat transport is investigated in the fascinating research field of spin caloritronics, the marriage of spintronics and thermoelectrics. Here, many new spin-dependent thermal transport phenomena in magnetic nanostructures have been explored in the recent years. One of them is the tunnel magneto-Seebeck (TMS) effect in magnetic tunnel junctions (MTJs) that has large potential for future nanoelectronic devices, such as nanostructured sensors for three-dimensional thermal gradients, or scanning tunneling microscopes driven by temperature differences. The TMS describes the dependence of the MTJ’s thermopower on its magnetic configuration when a thermal gradient is applied. In this review, we highlight the successes from the first observation of the TMS in 2011 to the current ongoing developments in this research area. We emphasize the different heating techniques, material designs, applications and additional physical aspects such as the role of the thermal conductivity of the barrier material. We further demonstrate the efficient interplay between ab initio calculations and experiments within this field, as this has led, e.g. to the detection of large TMS ratios in MTJs with half-metallic Heusler electrodes.

Keywords: tunnel magneto-Seebeck effect, magnetothermopower, magnetic tunnel junctions, tunnel magnetoresistance

(Some figures may appear in colour only in the online journal)

1. General introduction
In the emerging field of spin caloritronics [1–4], a huge amount of new spintronic effects related to thermoelectrics has been explored. The heart of spin caloritronics is the generation of spin-dependent phenomena by thermal means. Analog to the classical Seebeck effect [5], which is the generation of a thermoelectric voltage by the application of a temperature gradient, the spin-dependent Seebeck effect in magnetic metals and nanostructures is the thermovoltage (or thermocurrent) that depends on the magnetic state of the material or nanodevice. Here, the initial experiments [6–8] that started the spin caloritronics research field have been triggered by early magnetothermopower experiments in multilayers [9, 10]. One key spin-dependent phenomenon is the tunnel magneto-Seebeck (TMS) effect that describes the induced magnetothermopower in a magnetic tunnel junction (MTJ) (see figure 1(b)). The TMS was theoretically predicted by Czerner et al [11] and experimentally observed independently by Walter et al [12] as well as by Liebing et al [13] for MgO-based MTJs in 2011.
The first large effect in AlOx-based MTJs was reported by Lin et al [14].

This fascinating spin-dependent phenomenon is the spin caloritronic analog to the tunnel magnetoresistance (TMR) that has now been explored for decades in spintronics [15] (see figure 1(a)). Although both TMR and TMS have their origins in the spin-dependent transmission function of the MTJ and, therefore, are related to the spin-split density of states (DOS) of the involved materials, the magnitudes of both effects have no direct connection. While the TMR scales with the imbalance of majority and minority spin states close to the Fermi level, the TMS is affected by the spin-dependent asymmetry of the DOS with respect to the Fermi energy. This theoretical description has recently been proven experimentally by detecting a large TMS in MTJs with half-metallic Heusler alloys that have the Fermi energy close to one edge of the half-metallic band gap, thus supporting the TMS by a large asymmetry of the DOS with respect to the Fermi energy [16].

The inverse effect of the TMS, the tunnel magneto-Peltier (TMP) effect has been discovered already. It describes the generation of a spin-dependent temperature gradient across an MTJ when a charge current is applied [17] (see figure 1(c)). This effect together with the TMS effect fulfills the Onsager reciprocity [18] for spin-heat transport conversion. Parallel to the first TMS experiments in 2011, thermal spin injection into semiconductors, also known as Seebeck spin tunneling, was discovered [19–22].

The TMS in combination with a spin-transfer torque (STT) [23] has the potential to thermally assist the magnetic switching of MTJs [24]. After the first theoretical descriptions of the thermal STT in spin valves [25, 26] and MTJs [27, 28], the first evidence of an experimental confirmation can be found in [29–33]. While the electrically induced STT (magnetic switching by spin-polarized charge currents) is already utilized in commercially available STT-based magnetoresistive random-access memory devices, the thermal STT is not yet explored in a way that it can be used for commercial devices. Different applications are currently more promising such as three-dimensional sensing of thermal gradients [34] or scanning tunneling microscopy (STM) with temperature differences [35, 36] as presented in the following.

Within this review, we want to illustrate the TMS from first observation over improvement of devices and TMS magnitudes to future applications. In addition, we discuss fundamental open questions and ongoing research such as the accurate determination of the thermal gradient in the MTJs and the role of the thermal conductivity of the insulating barrier. This review starts with an introduction into the topic of TMR and TMS (section 2) including the description of typical TMS experiments subdivided by the individual heating techniques (laser, electric and intrinsic heating). Section 3 demonstrates that one can reach 3000% TMS by applying a bias voltage in addition to the temperature gradient. It further discusses different electrode and barrier materials, for example MgAl2O4, to optimize the TMS output. The final section, section 4, presents further aspects of the TMS, such as its angular dependence, the determination of the thermal conductivity of the tunnel barrier material and additional (magneto-)thermo-electric effects that have to be taken into account. We finally discuss in that section the influence of three-dimensional thermal gradients and the use of temperature differences in STM measurements.

2. Introduction to TMR and TMS

MTJs have always been important building blocks in spin- or magnetoelectronics [37]. The interest started with the discovery of TMR at room temperature in 1995 [38, 39]. Now, MTJs are used in magnetic logic devices, magnetic sensors and magnetoresistive random-access memories [37, 40]. The prediction [41, 42] and experimental realization [43, 44] of giant TMR in MgO-based MTJs as well as current-induced magnetic switching (caused by STT) in 2004 [45] further stimulated worldwide research activities.

An MTJ consists of a trilayer, in which two ferromagnetic electrodes are separated by a thin insulating tunnel barrier. A bias voltage applied across the tunnel barrier results in a current perpendicular to the layer plane [46]. A schematic of an MTJ is given in figure 1 for the individual spin-dependent effects mentioned so far.

The two extreme configurations of the electrodes’ magnetization are parallel (P) and antiparallel (AP) alignment. In most cases, the tunnel current is smaller in the AP than in the P case (positive TMR). However, for some materials the opposite case can occur which is related to a negative TMR.
The size of the TMR is connected to the spin polarization of the two electrodes and can be described by the model of Julliérc-model, which therefore cannot be used in the case of MTJs with symmetry-filtering properties.

If we exchange the applied bias voltage by a temperature gradient and replace the measured current by the thermovoltage as theoretically suggested in early literature [11, 52–54], we can observe the TMS [12, 13] as displayed in figure 1(b). The TMS depends on the magnetic configuration of the MTJ as the TMR does. However, the full magnetic field angular dependence is different as we will discuss in section 4.1. Analog to the TMR, we define the TMS as

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

Here, $S_P$ and $S_{AP}$ are the Seebeck coefficients for P and AP magnetic orientation of the MTJ. In contrast to the electric conductivity, the Seebeck coefficients can generally change sign. Hence, the TMS can have divergences whenever one of the Seebeck coefficients is zero [11].

If the transmission functions $T_P(E)$ and $T_{AP}(E)$ for the two magnetic configurations and the occupation function $f(E, \mu, T)$ are known, the Seebeck coefficients $S_P$ and $S_{AP}$ can be calculated via the moments $L_n$ obtained from the Landauer formalism [11, 55]. These moments are defined as

$$L_n = \frac{2}{\hbar} \int T(E)(E - \mu)^n(-\partial_E f(E, \mu, T))dE,$$

which are different for the P and AP case. The conductances $g_P$ and $g_{AP}$ as well as the Seebeck coefficients $S_P$ and $S_{AP}$ can be derived as
\[ g = \frac{e^2}{\hbar} L_0 \quad \text{and} \quad S = -\frac{1}{eT} \frac{L_1}{L_0}. \]  

(5)

From these equations one can conclude that the conductance \( g \) is proportional to the integral of the function

\[ T(E) \cdot \left( -\frac{\partial f(E)}{\partial E} \right), \]

(6)

whereas the Seebeck coefficient \( S \) is proportional to the center of mass of this function. Since \( g \) and \( S \) are not directly related, the TMR and TMS do not have a direct connection. However, they are determined by the same \( T(E) \) and, thus, are based on the same spin-split DOS of the materials involved.

For the theoretical approach, \textit{ab initio} calculations based on density-functional theory (DFT) have been conducted. In particular, the Korringa–Kohn–Rostoker (KKR) and the non-equilibrium Green’s function method have been used to obtain the transmission function \( T(E) \) [11] that leads to the conductance and Seebeck coefficients via equations (4) and (5) as well as to the TMR and TMS values via equations (1) and (3). For modeling the electrode material at the interface to the tunnel barrier, a supercell approach was applied in the beginning [11, 12, 56]. In subsequent studies the coherent potential approximation was employed in the KKR method [57], which has less computational effort and the possibility to use an arbitrary composition of the leads in contrast to the supercell approach. It turned out that non-equilibrium vertex corrections have to be taken into account within the KKR code [58]. Further theoretical descriptions and aspects can be found in the mentioned references summarized in a recent review about spin caloritric transport from DFT by Popescu et al [59].

In the following we will discuss the TMS for different kinds of heating techniques. The initial TMS observation in MTJs was achieved simultaneously by two groups. While Walter et al [12] used a laser to heat the top part of the MTJ, Liebing et al [13] patterned nanosized heater wires on top of the MTJ. Later on, Zhang and Teixeira et al [60, 61] presented an intrinsic heating approach using the applied tunnel current itself that creates a thermal gradient and thus an intrinsic TMS. This latter approach was discussed by Huebner et al [62] and compared to laser heating-induced TMS as recapped in section 2.3 of this review.

2.1 Laser heating-induced TMS

In the first laser heating-induced TMS experiments, a 784 nm laser was used with a focus of 15 \( \mu \text{m} \)–20 \( \mu \text{m} \) and a power of up to 100 mW to heat up the top part of the MTJ and, thus, to generate the temperature gradient [12]. In later experiments different laser wavelengths (e.g. 638 nm), smaller focus down to 2 \( \mu \text{m} \) and larger powers of up to 150 mW were employed [16, 63–65]. The Co–Fe–B/MgO/Co–Fe–B MTJs with a size of 1 \( \mu \text{m} \)–12.5 \( \mu \text{m} \) were patterned by electron beam lithography and ion beam etching as well as post-annealing [12].

A typical TMR and TMS result can be found in figures 2(a) and (b), respectively. The arrows indicate the magnetic alignment of the electrodes of the MTJ. The TMR curve shows a hard–soft switching with a positive TMR and 150% effect, while the TMS for a laser power of 30 mW loop has a drop in Seebeck voltage of \(-8.8\%\) going from 5.7 mV (P) to 5.3 mV (AP) thermovoltage.

In order to estimate the temperature difference across the barrier, finite element modeling based on the experimental parameters (laser focus and power, material properties such as the thermal conductivity) has been done as described in [12]. Thermal interface resistances and related effects have been neglected so far, but will be discussed in section 4.2 of this review. A temperature difference of \( \Delta T = 53 \text{ mK} \) has been derived for a laser focus of 15 \( \mu \text{m} \), a laser power of 30 mW and an MgO barrier thickness of 2.1 nm using the thin film value of \( \kappa = 4 \text{ W mK}^{-1} \) for the thermal conductivity of the MgO barrier [12]. The resulting \( S_P = V_P/\Delta T = -107.9 \mu \text{V K}^{-1} \) and \( S_{AP} = V_{AP}/\Delta T = -99.2 \mu \text{V K}^{-1} \) lead to the TMS of \(-8.8\%\) following equation (3).

Another way to determine the thermoelectric properties of these devices is to measure the thermoelectrically generated current of the device by a current amplifier in closed circuit conditions. A typical measurement of the laser heating-induced TMS current is shown in figure 3(b) and can be compared to the laser heating-induced TMS voltage via the known tunnel resistance (figure 3(a)). Note that the TMS shows an increase of the Seebeck current in the parallel state opposite to the Seebeck voltage behavior.

In general, the position and size of the laser strongly affects the TMS signal [63, 65]. It was found that the TMS vanishes for laser positions far off the centered position on the MTJ and maximizes if aligned at the center. For slight off-positions at the edges of the MTJs, additional heat transport effects such as the anomalous Nernst effect arise [34]. They will be discussed in section 4.3 of this review. The temperature dependence of the TMS has only been rarely studied so far [12]. Theoretical calculations have been carried out and sign changes of the TMS at specific temperatures depending on the Co–Fe interface termination [56] and composition [57] have been discussed. Systematic experimental temperature-dependent studies still have to be done to confirm the theoretical predictions.

In the beginning, only MTJs with an MgO tunnel barrier have been explored. In 2012, Lin et al reported on laser heating-induced TMS studies in MTJs with an aluminum oxide barrier [14] with similar values for the TMR and TMS of about 40%. The TMS is mostly independent from the heating power comparable to the results for MgO-barrier MTJs [13]. Furthermore, Lin et al [14] were able to invert the thermal gradient by heating either the top or bottom contact of the MTJ, thus inverting the detected thermocurrent. However, they could not observe any magnetic field dependence for the thermocurrent. This can be expected if the larger thermovoltage in the AP case is compensated by the simultaneously increased tunnel resistance. Later on, further theoretical studies on MTJs with an aluminum oxide barrier were conducted by López-Monis et al [66]. Huebner et al introduced MgAl2O4 tunnel barriers for laser heating-induced TMS investigations [62, 67, 68]. This will be further discussed in sections 2.3 and 3.2 of this review.
Most laser heating-induced TMS studies so far concentrate on MTJs with in-plane magnetized electrodes. However, some rare experiments have been done with MTJs exhibiting a perpendicular magnetic anisotropy (PMA). This type of MTJ is also interesting for STT switching experiments, since it usually needs less current density to manipulate the magnetization of one electrode by the spin-polarized current of the other. Thus, MTJs with PMA are also promising candidates for thermal STT experiments [30]. A typical MTJ stack with very thin barrier and electrode thicknesses supporting PMA is shown in figure 4(a). The resulting TMR (64.4%) and TMS (6%) curves are presented in figures 4(b) and (c), respectively. This kind of sample has been studied in the work of Leutenantsmeyer et al [30].

2.2. Electric heating-induced TMS

Another way to generate thermal gradients across the layer stack of an MTJ is the use of electric Joule heater lines (HL) lithographically placed on top of the MTJs. Electric heater schemes are well established to characterize in-plane thermoelectric properties of thin films [69]. Such electric heaters on membrane-based thermoelectric measurement platforms were used, e.g. to characterize the thermoelectric properties of magnetic thin films [70]. With respect to spin-dependent perpendicular transport electric heating was employed to characterize Seebeck spin tunneling from a ferromagnet into a semiconductor [19, 21] and spin heat accumulation in nanopillar spin valves [71]. Such electric heater schemes have also been realized by Liebing et al to study the TMS by detecting the tunnel magnetothermopower [13] and tunnel magnetothermocurrent [72] of Co–Fe–B/MgO/Co–Fe–B MTJ nanopillars.

In the first electric heater-induced TMS studies with in-plane magnetization, the MTJ stacks comprised a complex stack sequence including an antiferromagnetic pinning layer and a compensated synthetic antiferromagnetic reference layer [73]. Nanopatterning of the MTJ cells down to about 160 nm as well as contact definition was realized by electron beam lithography and clean room processing. A 5 µm wide HL was positioned on top of the nanopillar and separated from the MTJ top contact by a 160 nm thick Ta2O5 dielectric. Thermal gradients across the MTJ were generated by applying AC or DC heater currents of up to $I_{\text{heat}} = 60$ mA through the HL. Note that for electric heating the typical HL Oersted field of the order of 0.1 mT mA$^{-1}$ must be considered for data analysis. The heater temperature during the experiments can be determined by calibrating the temperature-dependent resistance change on a variable temperature probe station. This heater temperature and the temperature of the sample stage as input for finite element modeling of the heat flux through the complex layer stack can be utilized to estimate the temperature drop across the MgO barrier [74]. In the above experiments, maximum temperature drops of $\Delta T \approx 45$ mK were obtained.

Figure 2(d) shows typical thermopower data of these MTJ nanopillars. The measured thermopower voltage $V$ in open circuit conditions between the top and bottom contact of the MTJ is displayed for heater powers $P_{\text{heat}}$ of 21, 38 and 58 mW as a function of the magnetic easy axis field $H$. For all three curves $V_p$ in the P state is lower than $V_{\text{AP}}$ in the AP state with a maximum difference of $\Delta V \approx 11$ µV. One can identify a TMS value independent from the heating power with an average $\Delta V V_{\text{P}} \approx 32\%$. The black dashed line in figure 2(d) indicates the thermopower of the same MTJ patterned into nanopillars with diameter down to 160 nm after the MgO barrier was
applied to current stress (dielectric breakdown). The magnetic field-independent $V_{\text{short}}$ allows the determination of the thermopower contribution of all non-magnetic layers of the devices. Subtracting this background yields the true TMS value of the Co–Fe–B/MgO/Co–Fe–B MTJ reaching up to 90% for the given samples.

The different TMS magnitudes and the opposite signs between laser- and electric-heating TMS detection (Liebing et al [12]) for MgO-based in-plane magnetized MTJs can be explained by slight differences in the Co–Fe interface termination [56] and composition [57] of the MTJs. The theoretical work of Czerner and Heiliger shows that the influence of these electrode properties can have a huge impact on the TMS magnitude and sign. However, systematic experimental investigations varying only the interface termination or material composition while keeping the residual parameters of the MTJs constant is very challenging and could not be realized experimentally so far.

A typical example of the electric heating-induced TMS current is shown in figure 3(c). The experimental data can be well modeled taking into account the Onsager transport equations and the TMR of the MTJ devices [75, 76]. The electric current $I$ in the presence of the voltage $V$ and a temperature gradient $\nabla T$ across the MTJ is given by $I = gV - gS\nabla T$ where $g$ is again the electric conductance of the MTJ and $S$ the Seebeck coefficient. Measurements of the junction resistance $R$ as well as $V$ and $I$ in an open circuit ($I = 0$) and closed circuit ($V = 0$) configuration, respectively, allow the determination of $g = R^{-1}$ and $V = S\nabla T$. Using these values, the predicted magnetic thermocurrent $I$ in closed circuit configuration is given by $I = \frac{V}{R}$ and shows a good agreement with the experimental data (solid lines in figure 3(c)).

Böhnert et al extended these studies by the introduction of integrated thermometers allowing a better determination of the thermal gradient [77] and a more detailed analysis of the thermal interface transport (see section 4.2). They further varied the MgO thickness [78, 79] as summarized in section 3.3 of this review. So far, no electric heater-induced TMS experiments on MTJs with PMA have been conducted.

2.3. Intrinsic TMS

Besides laser and electric heating another method for the generation of a temperature gradient across an MTJ has been suggested by Zhang and Teixeira et al [60, 61], namely intrinsic heating by the tunnel current itself leading to an intrinsic TMS. In their articles, they explain that the tunnel current $I$ induces the thermal gradient via Joule heating and describe the resulting intrinsic thermopower effect as a nonlinear correction to Ohm’s law. They write

$$V(I) = RI + S\alpha I^2$$

with the resistance $R$ and the Seebeck coefficient $S$ of the MTJ as well as $\alpha = \sum_i \eta_i R_i R_{\text{MgO}}$. Here, $\eta_i$ is the thermal asymmetric parameter [60], $R_i$ the resistance and $R_{\text{MgO}}$ the thermal resistance of the $i$th layer. Thus, it is assumed that any quadratic-in-$I$ contribution in the $V-I$ curve of the MTJ can be related to this higher order term induced by a heating effect that modifies the measured voltage by an intrinsic TMS. However, this approach neglects any nonlinearity from the MTJ characteristics itself, e.g. barrier potential asymmetries.

In order to extract the intrinsic magneto-Seebeck coefficients from the $V-I$ curve for P and AP alignment, the data is separated into odd $((V_+ - V_-)/2 \propto I)$ and even $((V_+ + V_-)/2 \propto I)$ parts plotted against $I$ and $I^2$, respectively. The slope of the linear regressions (see equation (7)) gives the intrinsic magneto-Seebeck coefficients if $\alpha$ is estimated by a numerical calculation of the thermal profile of the MTJ [60–62].

With this approach Zhang and Teixeira et al concluded to have TMS values of more than 1000%. Ning et al [80] adopted this procedure and concluded to have similar large intrinsic TMS magnitudes for MTJs with PMA.

In order to compare the intrinsic TMS obtained by this symmetry analysis to the laser heating-induced TMS, Huebner et al [62] followed the same procedure for MgO and MgAl2O4 (MAO)-based MTJs. It turned out that in their case, the separation of odd and even parts of the $V-I$ curves did not necessarily give linear dependencies on $I$ and $I^2$, respectively. Therefore, a linear regression could only be made for small tunnel currents. The Seebeck coefficients obtained for the intrinsic TMS differed from the laser heating-induced Seebeck coefficients by more than one order of magnitude and had a different sign. As an example for MgO-MTJs, they determined $S_P = 0.3 \mu$V K$^{-1}$ and $S_{\text{AP}} = -7.5 \mu$V K$^{-1}$ for the intrinsic TMS via the symmetry analysis, while the laser heating-induced TMS resulted in $S_P = -1010 \mu$V K$^{-1}$ and $S_{\text{AP}} = -1320 \mu$V K$^{-1}$. Besides the quite different Seebeck coefficients, the intrinsic TMS values of 105% and $-75\%$ for MgO and MAO-based MTJs varied as well, compared to the laser heating-induced TMS ratios of 23% and 3.3% measured on the same MTJs, respectively.

Alternatively, the higher orders in the $V-I$ curve can be described by an asymmetric barrier potential without any connection to thermoelectrics. Using the Brinkman model [81], one can fit the curves varying the potential height $\varphi$, asymmetry $\Delta\varphi$ and thickness $d$ of the barrier. The model is valid

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**Figure 4.** Laser heating-induced TMS voltage for an MTJ with PMA. (a) Schematic of the MTJ’s structure. (b) TMR magnetic field loop. (c) TMS voltage loop for a laser power of 100 mW.
for MTJs with low or vanishing symmetry filter effect (constant transmission probability for all electrons). As discussed by Huebner et al., for this kind of MTJ the Brinkman model alternatively explains the higher orders in the $V–I$ curves with reasonable parameters [62]. They conclude that the intrinsic TMS cannot be unambiguously identified by the symmetry analysis proposed by Zhang et al because of the higher-order terms in the $V–I$ curve that are already present due to the barrier potential of the MTJ itself. Additional techniques and analyses have to be developed in the future to clearly distinguish between $V–I$ characteristics from any intrinsic TMS contribution and from the barrier potential of the MTJ.

3. Increase of TMS ratio and thermopower

There have been several ideas to increase the TMS magnitude. This includes the theoretical variation and optimization of the interface termination [56] and alloy composition [57] in case of Fe–Co electrodes. The latter basically shifts the Fermi level. Therefore, an idea arose from the experiment that the same goal could be reached by applying a bias voltage [64]. This bias-enhanced TMS has been studied theoretically and experimentally and will be discussed next, followed by further material optimization aspects such as the use of half-metal Heusler compounds as electrodes [64] or varying the thickness and material of the tunnel barrier [67, 78, 79].

3.1. Bias-enhanced TMS

The idea of shifting the Fermi level by a bias voltage is triggered by typical STM experiments where the electronic states can be selected by a bias voltage. In the case of the TMS it is more complicated, because there is not only the bias voltage but also the temperature gradient. Furthermore, because now a bias voltage is applied the thermopower voltage due to the temperature gradient cannot be measured directly. To overcome this issue the thermocurrent rather than the thermovoltage is detected as discussed in sections 2.1 and 2.2 of this review, but with and without the temperature gradient. Applying a temperature gradient not only leads to a temperature drop across the MTJ but also to an overall increase of average temperature. This is illustrated in figure 5.

Thereby, the mean temperature $\bar{T} = (T_L + T_R)/2$ changes by $\Delta \bar{T}$ whereas the temperature difference across the barrier is $\Delta T = T_L - T_R$ for $T_L$ and $T_R$ being the temperature at the upper and lower interface of the barrier, respectively (see figure 5). As shown in the examples of sections 2.1 and 2.2, simulations with finite element methods show that $\Delta T$ is in the order of 50 mK (for the given sample properties and heating parameters) and $\Delta \bar{T}$ can be several Kelvin depending on the time after the heating pulse [12].

In any case the Seebeck coefficient is no longer well defined and the current $I$ has to be considered, which is given by

$$I(\bar{T}, \Delta T, V_B) = \frac{2e}{h} \int dE T(E, V_B, \bar{T}) \left( f(E, \mu_L, \bar{T} - \frac{\Delta T}{2}) - f(E, \mu_R, \bar{T} + \frac{\Delta T}{2}) \right),$$

where $V_B = \frac{\mu_L - \mu_R}{e}$ is the applied bias voltage. Here, a cold current $I_c$ can be defined without any heating by using the occupation function

$$\Delta f_c = f(E, \mu_L, \bar{T}) - f(E, \mu_R, \bar{T}).$$

In the same way a hot current $I_h$ with heating is introduced by using the occupation function

$$\Delta f_h = f(E, \mu_L, T_L) - f(E, \mu_R, T_L).$$

In the experiment one measures the difference between both currents

$$\Delta I = I_h - I_c = \frac{2e}{h} \int dE T(E, V_B, \bar{T}) (\Delta f_h - \Delta f_c).$$

In order to understand which states are contributing to the detected current, $\Delta f_c - \Delta f_h$ should be analyzed. The principle is sketched in figure 6. In all cases one indeed gets contributions from other states but they are from the lower and the upper limit of the bias voltage window. In the case of a pure temperature gradient ($\Delta T = 0$) the difference in the occupation function is anti-symmetric. The latter is symmetric in the case of a pure increase of the average temperature ($\Delta \bar{T} = 0$). In reality it is a mixture of both cases. For example, in the experiment by Boehnke et al [64] finite element modeling shows that $\Delta T \gg \Delta \bar{T}$ as already mentioned above.

In order to get an idea about this effect figure 7(a) shows a realistic transmission function. From that one can calculate different scenarios using equation (11) for P and AP alignment. The bias-enhanced TMS can be defined by

$$\text{bTMS} = \frac{\Delta I_B - \Delta I_{AP}}{\min(|\Delta I_B|, |\Delta I_{AP}|)}.$$

The bTMS is shown for one scenario in figure 7(b). It is obvious that the bTMS can be easily tuned by the applied bias voltage. All this discussion neglects the influence of bias voltage and temperature on the transmission function. Therefore, this is a rather rough estimate and just gives an idea about the effect.

Corresponding measurements for the case that $\Delta \bar{T} \gg \Delta T$ are given by Boehnke et al [64] using laser heating. As presented in figure 8(b), the bTMS can reach values up to 3000%,
although the TMR is not that high (figure 8(a)). By varying the bias voltage the $\Delta T$ can reach zero for one magnetic configuration (see figure 8(c)). This zero crossing of one Seebeck coefficient in the denominator of the definition of equation (12) explains the high TMS ratios as presented in figure 8(b), for example. These divergences are also observable in the theoretic calculations and can be compared with the experimental values (figure 8(d)). For practical applications the absolute voltage value of $S_P$ or $S_{AP}$ has to be kept at a reasonable signal amplitude at the same time in order to exploit these large effects with a low noise level.

### 3.2. Variation of electrode material

The TMS ratios observed so far do not extend a few 10%, if we do not consider any intrinsic TMS and the bTMS. In order to increase the TMS by the choice of proper materials, one should take into account the dependence of the TMS on the asymmetry of the spin-split DOS with respect to the Fermi energy. Thereby, one has to consider two different cases. One goal might be to have large Seebeck values for parallel and anti-parallel alignment but with opposite sign. Such transmission functions are shown in figure 9(a). In this case the TMS ratio as defined by equation (3) might be not that high.

Therefore, another idea is to have a very small Seebeck coefficient for one alignment and a large one for the other alignment. This situation is shown in figure 9(b). (b) One can discuss how an MTJ with such transmission functions should be designed. A simple way to answer this question is to apply the Juliére model [15], which connects in a simple way the available DOS $n$ to the transmission function via

$$T_P \propto n_L \cdot n_R \uparrow + n_L \cdot n_R \downarrow,$$

$$T_{AP} \propto n_L \cdot n_R \downarrow + n_L \cdot n_R \uparrow,$$  

(13)

where $L$ and $R$ again refer to the material above and below the barrier, respectively. The Juliére model is valid only for incoherent tunneling but it gives some clue as to what kind of material is necessary. Applying the Juliére model to the transmission function shown in figure 9(a) leads to the finding that one magnetic layer has to be a half-metal whereas the other one could be a magnetic semiconductor with properly aligned bands. This is shown in figure 9(c).

For the other case a possible solution is to have one half-metallic magnetic layer and a second half-metallic layer with properly aligned bands that show a peak in one spin channel close to the Fermi level. Indeed such a DOS is common for certain Heusler compounds [16, 82, 83]. A similar Heusler on the other side of the barrier would not get the desired behavior, because the two peaks would lead to an asymmetric transmission function in the AP case and, thus, to a non-vanishing Seebeck coefficient. In the case of the epitaxial MgO-based MTJs it is very simple to get the half-metallic properties, since the MgO/Fe or MgO/Fe–Co interface acts for the transport the MgO/Fe or MgO/Fe–Co. Following this idea, Boehnke et al [16] experimentally investigated MTJs with the half-metallic Heusler compounds Co$_2$FeAl and Co$_2$FeSi. As an example, we have picked the case of MTJs with one Co$_2$FeAl electrode to illustrate this successful path from material design to high effect values. The DOS of Co$_2$FeAl in the B2 structure calculated by DFT is shown in figure 10(a).

One can clearly see that the Fermi level is located at the edge of the half-metallic band gap of Co$_2$FeAl in the B2 structure. This is also the case for Co$_2$FeSi in the L2$_1$ structure [16]. For L2$_1$-structured Co$_2$FeAl, the Fermi level is in the center of the band gap. That is why B2-structured Co$_2$FeAl is more
likely to give a large TMS. Boehnke et al prepared Co2FeAl MTJs with a Co–Fe–B counter-electrode and compared the TMS as well as the TMR with MTJs having both electrodes made of Co–Fe–B. The results for the laser heating-induced TMS are presented in figure 10(b). While the TMS ratio of the Co–Fe–B MTJ stays below 50%, the TMS ratio of the Co2FeAl MTJ nearly exceeds $-100\%$. This result confirms the predictions made based on the calculated DOS and the position of the Fermi energy (figure 10(a)) as well as the considerations made with respect to the desired transmission functions (figure 9).

If we further compare the TMR ratios of the two types of MTJs (figure 10(c)), we identify a smaller TMR for the Co2FeAl MTJ compared to the Co–Fe–B MTJ. This again confirms the fact that the TMS and TMR are not directly related. Boehnke et al investigated multiple MTJs of each kind and also varied the Co–Fe composition in agreement with the theoretical predictions of Heiliger et al [57]. However, the same trend was observed for all samples: while large Seebeck coefficients in combination with large TMS values up to $-120\%$ have been observed for the Heusler MTJs, the values are much smaller for the Co–Fe–B MTJs. This result is independent from the magnitude of the TMR ratio as systematically studied by Boehnke et al [16].

Further material variations have been calculated by Amin et al [85] by taking advantage of materials with large spin–orbit coupling, such as Pt. They calculated a large tunnel anisotropy magneto-Seebeck effect in CoPt/MgO/Pt tunnel junctions of 175%.

3.3. Variation of barrier material and thickness

The thickness of the insulating barrier and the choice of the barrier material play another important role. This has been systematically varied experimentally by Huebner et al [67] as well as Böhner et al [78, 79]. Besides MgO [12, 13] and Al2O3 [14] as barrier materials, the spinel ferrite MAO is an interesting candidate with fascinating properties. MAO has a lower lattice mismatch with standard ferromagnetic electrodes (Fe, CoFe, CoFeB, etc) of 1% compared to MgO that has between 3% and 5% [86]. This low value can be even more reduced by growing MgAl2O4 with reduced oxygen content via molecular beam epitaxy [87]. As a tunnel barrier, MAO exhibits a similar symmetry filter effect as MgO [88]. However, experimental TMR ratios obtained so far are still below the values for MgO-based MTJs [89–91], while magnetization switching by STT has been demonstrated [92] and MgAl2O4 double-barrier systems with pronounced resonant tunneling features in quantum well structures have been realized [87].
In figure 11, the TMR and TMS ratios together with the thermovoltage for MgO and MAO barrier MTJs with Co–Fe–B electrodes are plotted against the resistance area product \( RA \) that scales with the barrier thickness [67]. The barrier thicknesses from 1 nm to 3 nm result in resistance area products in the range of \( 10^{-1} \) to \( 10^3 \) k\( \Omega \) m\(^2\). For each barrier thickness several MTJs are characterized and averaged. The TMR is maximal at around a nominal barrier thickness of 2 nm. Values up to 30% for MAO-based MTJs with \( RA = 100 \) k\( \Omega \) m\(^2\) and 150% for MgO-based MTJs with \( RA = 1000 \) k\( \Omega \) m\(^2\) can be observed (figure 11(a)).

If the TMS ratios between MAO- and MgO-based MTJs are compared, the TMS is again larger for MgO-based MTJs (figure 11(b)). The maximum is observed for both types of MTJs at a nominal barrier thickness of 2.6 nm (\( RA \) between 1000 k\( \Omega \) m\(^2\) and \( 10^4 \) k\( \Omega \) m\(^2\)). Experiments and theory do not agree here, since theory predicts an increasing TMS ratio when going down from ten monolayers (MLs) (2%) to six monolayers (10%) of MgO (1 ML = 2.1 Å) [57]. However, the interface structure can be quite different in the experiment compared to the theory, where it is assumed to be ordered perfectly. Further decrease of TMR and TMS values for thinner barriers is due to pinholes that become more probable for smaller thicknesses and reduce the effect amplitudes. Since the TMS is sensitive to the interface structure [56] and barrier properties, this can already lead to discrepancies between theory and experiment.

Beside large TMS ratios, a huge thermopower is desirable. Therefore, the thermopower is plotted against the barrier thickness, i.e. \( RA \) (figure 11(c)), for a laser power of 150 mW and MTJs with an area of \( 6\pi \) m\(^2\). Although the TMS is smaller in MAO-based MTJs compared to MgO-based ones, the thermopower is larger for the MAO barrier throughout most of the thicknesses tested. The highest value has been found for the thickest MAO barrier of 2.6 nm. The two samples with a nominal MAO thickness of 1.8 nm and 2.0 nm have been fabricated and measured separately under different experimental conditions which could explain the reduced thermopower values as discussed in [67]. The thermopower has been taken after subtracting the lead contribution determined after electric breakdown of the MTJ by applying 3 V to the junction.

In addition, Böhnert et al [78, 79] investigated electric heater-induced TMS in MTJs with Co–Fe–B electrodes and an MgO barrier wedge with varied thicknesses from 1.2 nm to 1.6 nm related to 0.7 k\( \Omega \) m\(^2\) and 55 k\( \Omega \) m\(^2\), respectively. The obtained TMS values between 5% and 35% are in a comparable order of magnitude to the results of Hübner et al [67]. Theoretical studies on MTJs with varying thickness of an aluminum oxide barrier have been conducted by López-Monís et al [66]. However, experimental investigations for this kind of MTJ with various thicknesses of the aluminum oxide barrier are still missing.

4. Further aspects and applications

4.1. Angular dependence of the TMS

Up to now we discussed the P and AP alignment of the magnetic moments. However, the magnetization of the two ferromagnetic layers can be oriented in any direction to each other. For the case of the TMR or the STT this gives basically a simple cosine dependence [23, 93, 94]. For the TMS this can be different [95]. Figure 12(a) shows the calculated normalized angular dependence for three different MTJs. The main difference between these junctions is the TMR ratio. It can be shown that the angular dependence of the Seebeck coefficient S follows [95]

\[
S(\theta) = \frac{S_P \cdot \text{TMR} + S_P + S_{AP} + (S_P \cdot \text{TMR} + S_P - S_{AP}) \cdot \cos(\theta)}{\text{TMR} + 2 + \text{TMR} \cdot \cos(\theta)}
\]

(14)

From that it becomes clear that in the limiting case of vanishing TMR the TMS obeys a cosine dependence. This also explains the different results between theory and experiments, because in the experiments TMR ratios are typically between 100% and 300% whereas in theory they are typically far above 1000%. In figure 12(b) we show the experimental results compared to equation (14) for different TMR ratios. One obtains a fairly good agreement with equation (14) by plugging in the TMR ratio of the experiment. The results show that although the TMS and TMR ratios are independent they are connected via the angular dependence. In particular, the TMR ratio can be fitted by applying equation (14) to the experimental data.

4.2. Temperature drop across a tunnel barrier

All the discussed experiments need a temperature gradient across an MTJ. Whereas in theory the transport parameters are in linear response, i.e. with a vanishing temperature gradient, in experiments one has a basically unknown temperature drop across the barrier. However, this drop is important to calculate for example the Seebeck coefficient, because only a thermovoltage is measured. Recently, there have been some
be dominated by electrons whereas the transport through the insulator will be dominated by phonons.

Therefore, Zhang et al [98] calculated the PDOS for an MTJ with three monolayers of MgO sandwiched between four monolayers of Fe using non-equilibrium Green’s functions. The structure together with the projected PDOS are shown in figure 13. In addition, the PDOS of bulk MgO and bulk Fe is also shown. By comparing these two it is obvious that there is a huge mismatch. In particular, there is a cut-off energy of about 40 meV given by Fe, and at the energy range from 20 meV to 30 meV where Fe has a rather high PDOS, MgO shows a very low PDOS. Such a mismatch means that there is a very high thermal interface resistance.

This is reflected in the corresponding transmission function shown in figure 14(a). Thereby, the transmission function is almost independent of the MgO thickness. From the transmission function one can calculate the thermal conductance shown in figure 14(b). These values are already orders of magnitude smaller than the bulk values of MgO [99] and even smaller than the values of MgO thin films [100]. For comparison figure 14(c) shows the electron thermal conductance. As expected it decays exponentially with the increasing MgO thickness. Even at thin MgO barriers it is one order of magnitude smaller than the phonon thermal conductance.

In order to get the thermal conductance of the whole MTJ the energy balance equations can be solved for the electron and phonon temperature, \( T_e \) and \( T_p \) [98, 101]

\[
\frac{k_e}{d} \frac{d^2T_e(z)}{dz^2} - G_{eph}(T_e - T_p) = 0,
\]

\[
\frac{k_p}{d} \frac{d^2T_p(z)}{dz^2} + G_{eph}(T_e - T_p) = 0,
\]

where \( z \) is the transport direction and \( G_{eph} \) is the electron–phonon interaction factor [102]. \( k_e \) and \( k_p \) are the electron and phonon thermal conductivities in Fe, respectively. The results are shown in figure 15.

It becomes clear that there is a huge temperature drop at the MgO barrier, which was already expected from figure 14. Furthermore, an imbalance of the electron and phonon temperature at the interface is visible. The reason is that the electrons get more reflected than the phonons at the interface and thus show a heat accumulation. The electron–phonon coupling leads to an equilibration of \( T_e \) and \( T_p \) away from the interface. The inset of figure 15 shows a fictitious case with an MTJ that has a ten times larger thermal conductance.

Although there is a non-equilibrium situation at the Fe/MgO interface one can estimate the total thermal conductance through the whole MgO stack [98]. These values as a function of MgO thickness are given in figure 16 at room temperature and can be used as input parameters in finite element methods.

In figure 16 these values are compared to values extracted from bulk MgO and thin-film MgO. Going from bulk to thin films the thermal conductance is already decreased by one order of magnitude but it is again reduced by one order of magnitude going to the MTJ. Thus the values used in the

![Figure 11](image-url)

**Figure 11.** Variation of the barrier thickness from 1 nm to 3 nm which affects the resistance area product \( R_A \) for MAO (left axis, blue circles) and MgO (right axis, red squares) barrier MTJs. (a) Comparison of the averaged TMR ratios over all measured elements plotted against \( R_A \). (b) Comparison of the laser heating-induced TMS ratios (averaged over all measured elements) plotted against \( R_A \). (c) Measured absolute thermopower with a laser power of 150 mW of the MTJs with an area of 6\( \mu m^2 \) plotted against \( R_A \). Reprinted figure with permission from [67]. Copyright 2017 by the American Physical Society.
Figure 12. Angular dependence of the TMS. (a) Seebeck coefficients obtained by ab initio calculations [11, 12, 57]. The symbols are the angles where the calculations have been done and the solid lines are equation (14). (b) Experimental data of the angular dependence of the Seebeck coefficient compared to equation (14) for different TMR ratios. Reproduced from [95], with the permission of AIP Publishing.

past for finite element methods [12] lead to an underestimation of the temperature drop and thus to an overestimation of the Seebeck coefficient. This finding is confirmed by recent experiments [68, 77].

4.3. Three-dimensional thermal gradients and additional (magneto-)thermoelectric effects

Additional (magneto-)thermoelectric effects can occur in TMS experiments depending on the used MTJ materials and the heating technique. Here, we will discuss two examples. If Si is used as a substrate for the MTJs, additional Seebeck voltages [63] and photocurrents [103] can be created in laser heating-induced TMS experiments depending on the spot size of the laser. Furthermore, unintended in-plane thermal gradients can end up as anomalous Nernst effect (ANE) contributions in the electrodes as systematically studied by Martens et al [34]. Since the heat dissipation of the laser spot occurs in all spatial directions, additional (magneto-)thermoelectric effects open new opportunities for the detection of the overall direction of the thermal gradient based on different heating scenarios and the control of the temperature gradient direction.

Figure 13. (a) The sketch of the Fe/MgO(3ML)/Fe MTJ for the calculation of phonon transmission function. The supercell used for the phonon calculations is marked as scattering region. (b) The corresponding total and projected phonon density of states (PDOS). In addition the bulk PDOS of Fe and MgO are shown. Reprinted figure with permission from [98], Copyright 2015 by the American Physical Society.

The lock-in detection of the laser heating-induced TMS experiments allows the time-resolved measurements of the TMS. Thus, any rising time of the thermovoltages and additional thermoelectric effects can be identified. By comparing the time-dependent TMS for MTJs on MgO and SiO2/Si substrates, Boehnke et al identified an additional Seebeck effect from the Si substrate that is not detectable in the case of MgO [63]. While for MgO the thermovoltage increases within hundreds of µs up to a plateau value, the thermovoltage of the Si samples overshoots this plateau value and decreases back down, probably due to additional capacitive couplings in the sample structure. Boehnke et al developed a model circuit that considers the additional thermovoltage from the Si together with a capacitive coupling through the SiO2 layer and could nicely describe the time-dependent experimental data.

The effect of a Seebeck voltage from the substrate is only pronounced if the laser spot is placed acentric on the MTJ [63]. This hints to the fact that additional in-plane thermal gradients induce the Seebeck effects of the substrate. As long as they are not canceled out (as is the case for the acentric position of the laser spot), the Seebeck voltage of the substrate is contributing. Since MgO is insulating compared to the semiconducting Si, no additional Seebeck effect can be observed for MgO substrates independent from the position of the laser spot.

Beside this thermoelectric effect from in-plane thermal gradients, Martens et al studied magnetothermoelectric effects, such as the ANE [104–106], for in-plane thermal gradients in the electrode material [34]. They are able to control the temperature gradient direction with respect to the magnetization alignment and extract the contribution of the ANE to the TMS signal. The measurements of TMS voltages in pseudo spin valves reveal a characteristic shift, (see figure 17(a)) for parallel magnetization alignment in opposite magnetic field directions.
The shift amplitude for each heating position is plotted in figure 17(b) in a three-dimensional plot and projected to the bottom of the graph with a black elliptically shaped contour line that indicates the MTJ position. The analysis of the extracted voltage shows a behavior that corresponds to the ANE. In particular, the voltages $\Delta V_{\text{ANE}}$ extracted at heating positions at the MTJ’s edge, marked by the contour (see figure 17(c)), can be described by a sinusoidal dependence of the magnetization direction pointing to the typical angular dependence of the ANE [34].

Future work could relate the individual effects to quantitative values that are reliable enough to compare the thermal gradient components of the TMS (out-of-plane component of $\nabla T$) and the ANE (in-plane component of $\nabla T$). Thus, one could receive the effective spatial direction of the thermal gradient and create a three-dimensional temperature gradient sensing device.

4.4. Vacuum gap as tunnel barrier

The TMS can be also observed on the atomic scale if a vacuum gap is used as the tunnel barrier [35, 36]. Friesen et al investigated STT and TMS on magnetic single and double layers using magnetic tips for STM. As depicted in figure 18(a), the tip is heated by a laser and, thus, creates the temperature gradient. The thermal expansion of the tip is used to determine quantitative numbers for the temperature difference. Due to this laser heating-induced thermal gradient the TMS can be detected as a thermovoltage and be mapped across the studied surface with atomic resolution for different magnetic moment alignments depending on the given sample.

Here, Friesen et al have studied the TMS in Fe single and double layers grown on W(110) and Ir(111) substrates [36]. They clearly observe a linear dependence of the temperature difference between tip and surface on the magnetothermopower. Mapping of the local compensation bias voltage $U_c$ of the thermopower for zero DC tunnel current leads to the spatially resolved Seebeck coefficient $S$. The compensation to zero tunnel current allows the exclusion of additional DC Joule heating effects. This measurement technique that is based on AC tunnel currents can even be applied for a vanishing temperature gradient, as discussed in [36]. In single layers of Fe on W(110) in-plane alignment of the magnetic moments is preferred while for double layers out-of-plane alignment is favorable and the moments align in magnetic domains and domain walls, which is also observable in the mapping of the Seebeck coefficient.

The Seebeck coefficients on the double layers can be described by a combination of spin-averaged Seebeck tunneling (ST), TMS and tunneling anisotropic magneto-Seebeck (TAMS) thermopower generated by spin–orbit coupling and by the different aligned magnetic moments within the magnetic domains and across the domain walls. The difference between three effects is shown in figure 18(b) and they have been separated in the work of Friesen et al [36]. As an example, the mapping of the spin-averaged ST response is shown in figure [35] with the $U_c$ values of the single and double layers for a Fe/W(110) sample. A standard tunnel current STM (I-STM) image of the same area is presented for comparison. The mapping of the magnetic response and additional results such as the mapping of $S$ for the Skyrmion material Fe/Ir(111) can be found in [35]. Generally, thermovoltages
in STM experiments show fine contrast and features of electronic states also in non-magnetic samples because of their different dependence on the local DOS as compared to an electrically biased tunneling.

5. Conclusion and outlook

The TMS in MTJs is a fascinating spin caloritronic effect that provides the possibility to obtain thermovoltages depending on the magnetic configuration of the MTJ and, thus, to use heat for future elements of spintronic circuits. We have reviewed the successes of the TMS from first detection by laser and electric heating in Co–Fe–B/MgO/Co–Fe–B MTJs [12, 13] over material optimization aspects to open questions that still have to be solved.

The experimental equipment and theoretical implementations are already at a very high level, so that ab initio calculations of band structure and transmission function nicely agree with the features that are observed in experiments. This scientific tango between theory and experiment [107] led, for example, to the prediction and experimental confirmation of high TMS ratios in MTJs with half-metallic Heusler electrodes [16]. We further presented the increase of thermovoltages for the use of MAO barriers with an optimized barrier thickness of 2.6 nm [67]. Future studies should be made for Heusler/MAO/Co–Fe–B MTJs to adjust both high TMS ratios and large thermovoltages.

Further aspects that have to be addressed are the disentanglement of any intrinsic TMS effect induced by the tunnel current itself from the characteristic properties of the barrier shape that influence the V–I curve in a similar way [62]. The role of the barrier thermal conductivity and additional thermal interface conductivities between barrier and electrodes are not yet fully explored. Due to the theoretical work done by Zhang et al [98] and experiments by Huebner et al [68] as well as Böhnt et al [77] barrier thermal conductivities at least one order of magnitude below the bulk value or even lower can be expected. Additional optical experiments based on time-domain thermoreflectance [108] could shine light on these buried parts of TMS research.

The TMS in double barriers [109, 110] and the role of magnon transport in the TMS [111] have been discussed recently from the theoretical side. However, experiments following these ideas have not yet been conducted. Finally, the community should move forward towards applications and explore devices that combine the TMS with other spintronic or (magnetoo-)thermoelectric effects to integrate MTJs in nano electronics while recovering unused heat in those nanostructures. Nanostructured sensing devices for three-dimensional thermal gradients [34] and the use of thermal gradients in STM devices [36] are only the first realizations of new TMS applications.
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References

[1] Bauer G E W, Saiot E and van Wees B J 2012 Spin caloritronics Nat. Mater. 11 391–9
[2] Münzenberg M and Thomas A 2012 Heiße Elektronik—Vom Seebeck-Effekt zur Spinkaloritronik Phys. Unserer Z. 43 288
[3] Boona S R, Myers R C and Heremans J P 2014 Spin caloritronics Energy Environ. Sci. 7 885–910
[4] Yu H, Brechet S D and Ansermet J-P 2017 Spin caloritronics, origin and outlook Phys. Lett. A 381 825–37
[5] Seebeck T 1822–1823 Magnetische Polarisation der Metalle und Erze durch Temperatur-Differenz (Abhandlungen der Preussischen Akademie der Wissenschaften) (Leipzig: W Engelmann) pp 265–373
[6] Gravier L, Serrano-Guisan S, Reuse F and Ansermet J-P 2006 Thermodynamic description of heat and spin transport in magnetic nanostructures Phys. Rev. B 73 024419
[7] Uchiha K, Takahashi S, Harii K, Ieda J, Koshiba W, Ando K, Maekawa S and Saitoh E 2008 Observation of the spin Seebeck effect Nature 455 778–81
[8] Slachter A, Bakker F L, Adam J P and van Wees B J 2010 Thermally driven spin injection from a ferromagnet into a non-magnetic metal Nat. Phys. 6 879–82
[9] Shi J, Parkin S S P, Xing L and Salamon M B 1993 Giant magnetoresistance and magnetothermopower in Co/Cu multilayers J. Magn. Magn. Mater. 125 L251
[10] Baiiya S A, Salamon M B and Oepts W 2000 Magnetothermopower of cobalt/copper multilayers with gradient perpendicular to planes J. Appl. Phys. 87 4855
[11] Czerner M, Bachmann M and Heiliger C 2011 Spin caloritronics in magnetic tunnel junctions: ab initio studies Phys. Rev. B 83 132405
[12] Walter M et al 2011 Seebeck effect in magnetic tunnel junctions Nat. Mater. 10 742–6
[13] Liebing N, Serrano-Guisan S, Rott K, Reiss G, Langer J, Ocker B and Schumacher H W 2011 Tunneling magnetothermopower in magnetic tunnel junction nanopillars Phys. Rev. Lett. 107 177201
[14] Lin W, Hehn M, Chaput L, Negulescu B, Andrieu S, Montaigne F and Margin S 2012 Giant spin-dependent thermoelectric effect in magnetic tunnel junctions Nat. Commun. 3 744
[15] Juliére M 1975 Phys. Lett. A 54 225
[16] Boehnke A et al 2017 Large magneto-Seebeck effect in magnetic tunnel junctions with half-metallic Heusler electrodes Nat. Commun. 8 1626
[17] Shan J, Dejene F K, Leutensmeyer J C, Flipse J, Münzenberg M and Van Wees B J 2015 Comparison of the magneto-Peltier and magneto-Seebeck effects in magnetic tunnel junctions Phys. Rev. B 92 020414
[18] Onsager L 1931 Reciprocal relations in irreversible processes. 1 Phys. Rev. 37 405–26
[19] Le Breton J-C, Sharma S, Saito H, Yuasa S and Jansen R 2011 Nature 475 82
[20] Jansen R 2012 Silicon spintronics Nat. Mater. 11 400–8
[21] Jeon K-R, Min B-C, Spiesser A, Saito H, Shin S-C, Yuasa S and Jansen R 2014 Voltage tuning of thermal spin current in ferromagnetic tunnel contacts to semiconductors Nat. Mater. 13 360
[22] Jeon K-R, Saito H, Yuasa S and Jansen R 2015 Relative strength of thermal and electrical spin currents in a ferromagnetic tunnel contact on a semiconductor Phys. Rev. B 92 054403
[23] Slonczewski J C 1989 Conductance and exchange coupling of two ferromagnets separated by a tunneling barrier Phys. Rev. B 39 6995
[24] Pushp A, Phung T, Rettner C, Hughes B P, Yang S-H and Parkin S P S 2015 Giant thermal spin-torqueassisted magnetic tunnel junction switching Proc. Natl Acad. Sci. USA 112 6585–90
[25] Hatami M, Bauer G E W, Zhang Q and Kelly P J 2007 Thermal spin-transfer torque in magnetoelectronic devices Phys. Rev. Lett. 99 066603
[26] Slonczewski J C 2010 Initiation of spin-transfer torque by thermal transport from magnons Phys. Rev. B 82 054403
[27] Jia X, Xia K and Bauer G E W 2011 Thermal spin transfer in Fe–MgO–Fe tunnel junctions Phys. Rev. Lett. 107 176603
[28] Heiliger C, Franz C and Czerner M 2014 Thermal spin-transfer torque in magnetic tunnel junctions (invited) J. Appl. Phys. 115 172614
[29] Yu H, Granville S, Yu D P and Ansermet J-P 2010 Evidence for thermal spin-transfer torque Phys. Rev. Lett. 104 146601
[30] Leutensmeyer J C et al 2013 Parameter space for thermal spin-transfer torque Spin 03 1350002
[31] Zhang Z, Bui L, Chen X, Guo H, Fan X L, Xue D S, Houssameddine D and Hu C-M 2016 Observation of thermal spin-transfer torque via ferromagnetic resonance in magnetic tunnel junctions Phys. Rev. B 94 064414
[32] Liebing N, Serrano-Guisan S, Rott K, Reiss G and Schumacher H 2016 Noise spectroscopy of CoFeB/MgO/CoFeB magnetic tunnel junctions in the presence of thermal gradients J. Magn. Magn. Mater. 400 154–8
[33] Cansever H, Raskovicz R, Lenz K, Fowley C, Ramasubramanian L, Yildirim O, Niesen A, Huebner T,
Reiss G and Lindner J 2018 Investigating spin-transfer torques induced by thermal gradients in magnetic tunnel junctions by using micro-cavity ferromagnetic resonance J. Phys.: D: Appl. Phys. 51 224009

Martens U et al 2018 Anomalous Nernst effect and three-dimensional temperature gradients in magnetic tunnel junctions Commun. Phys. 1 65

Friesen C, Osterhage H, Friedlein J, Schlenhoff A, Wiesendanger R and Krause S 2018 Scanning Sbeebeck tunneling microscopy J. Phys.: D: Appl. Phys. 51 324001

Friesen C, Osterhage H, Friedlein J, Schlenhoff A, Wiesendanger R and Krause S 2018 Magneto-Sbeebeck tunneling on the atomic scale Science accepted

Prinz G 1998 Magnetoelectronics Science 282 1660

Moodera J, Kinder L and Wong T 1995 Large magnetoresistance at room temperature in ferromagnetic thin film tunnel junctions Phys. Rev. Lett. 74 3273

Miyazaki T and Tezuka N 1995 Giant magnetic tunneling effect in Fe/Al2O3/Fe junction J. Magn. Magn. Mater. 139 I.231

Miao G-X, Münzenberg M and Moordera J S 2011 Tunneling path toward spintronics Rep. Prog. Phys. 74 036501

Mathon J and Umerski A 2001 Theory of tunneling magnetoresistance of an epitaxial Fe/MgO/Fe(001) junction Phys. Rev. B 63 224043

Butler W H, Zhang X-G and Schulthess T C 2001 Spin-dependent tunneling conductance of Fe/MgO/Fe sandwiches Phys. Rev. B 63 054416

Parkin S S P, Kaiser C, Panchula A, Rice P M, Hughes B, Samant M and Yang S-H 2004 Giant tunneling magnetoresistance at room temperature with MgO (100) tunnel barriers Nat. Mater. 3 862

Yuasa S, Nagahama T, Fukushima A, Suzuki Y and Ando K 2004 Giant room-temperature magnetoresistance in single-crystal Fe/MgO/MgFe magnetic tunnel junctions Nat. Mater. 3 868

Huang Y, Albert F, Nguyen P, Pakala M and Valet T 2004 Observation of spin-transfer switching in deep submicron-sized and low-resistance magnetic tunnel junctions Appl. Phys. Lett. 84 3118

Moordera J S and Mathon G 1999 Spin polarized tunneling in ferromagnetic junctions J. Magn. Magn. Mater. 200 248

Sharma M, Wang S X and Nickel J H 1999 Inversion of spin polarization and tunneling magnetoresistance in spin-dependent tunneling junctions Phys. Rev. Lett. 82 616

De Teresa J M, Barthlmy A, Bert A, Contour J P, Lyonnet R, Montaigne F, Seneor P and Vaurs A 1999 Inverse tunnel magnetoresistance in Co/STiO3/La0.7Sr0.3MnO3: new ideas on spin-polarized tunneling Phys. Rev. Lett. 82 4288

Kliewe C, Meinert M, Schmalhorst J and Reiss G 2013 Negative spin polarization of Mn3V2Ga probed by tunnel magnetoresistance J. Phys.: Condens. Matter 25 076001

Maranz L et al 2015 Sign change in the tunnel magnetoresistance of FeOx/MgO/CoFeB magnetic tunnel junctions depending on the annealing temperature and the interface treatment AIP Adv. 5 047103

Ikeda S, Hayakawa J, Ashizawa Y, Lee Y M, Miura K, Hasegawa K, Tsunoda M, Matsukura F and Ohno H 2008 Tunnel magnetoresistance of 604% at 300 K by suppression of Ta diffusion in CoFeB/MgO/CoFeB pseudo-spin-valves annealed at high temperature Appl. Phys. Lett. 93 082508

Wang Z-C, Su G and Gao S 2001 Spin-dependent thermal and electrical transport in a spin-valve system Phys. Rev. B 63 224419

McCann E and Falko V I 2002 Giant magnetothermopower of magnon-assisted transport in ferromagnetic tunnel junctions Phys. Rev. B 66 134424

McCann E and Falko V I 2002 Magnetothermopower and magnon-assisted transport in ferromagnetic tunnel junctions Appl. Phys. Lett. 81 3609

Ouyang Y and Guo J 2009 A theoretical study on thermoelectric properties of graphene nanoribbons Appl. Phys. Lett. 94 263107

Czerny M and Heiliger C 2012 Influence of interface termination on the magneto-Seebeck effect in MgO based tunnel junctions J. Appl. Phys. 111 07C511

Heiliger C, Franz C and Czerny M 2013 Ab initio studies of the tunneling magneto-Seebeck effect: influence of magnetic material Phys. Rev. B 87 224412

Franz C, Czerny M and Heiliger C 2013 Implementation of non-equilibrium vertex corrections in KKR: transport through disordered layers J. Phys.: Condens. Matter 25 425301

Popescu V et al 2018 Spin caloric transport from density-functional theory J. Phys. D: Appl. Phys. 52 073001

Zhang Z H et al 2012 Seebeck rectification enabled by intrinsic thermoelectric coupling in magnetic tunneling junctions Phys. Rev. Lett. 109 037206

Teixeira J M, Costa J D, Ventura J, Fernandez-Garcia M P, Azevedo J, Araujo J P, Sousa J B, Wisnikowski P, Cardoso S and Freitas P P 2013 Giant intrinsic thermoelectric effects in thin MgO magnetic tunnel junctions Appl. Phys. Lett. 102 212413

Huebner T, Boehnke A, Martens U, Thomas A, Schmalhorst J-M, Reiss G, Münzenberg M and Kuschel T 2016 Comparison of laser-induced and intrinsic tunnel magneto-Seebeck effect in CoFeB/MgAl2O4 and CoFeB/MgO magnetic tunnel junctions Phys. Rev. B 93 224433

Boehnke A, Walter M, Roschewsky N, Eggebrecht T, Drewello V, Rott K, Münzenberg M, Thomas A and Reiss G 2013 Time-resolved measurement of the tunnel magneto-Seebeck effect in a single magnetic tunnel junction Rev. Sci. Instrum. 84 063905

Boehnke A et al 2015 On/off switching of bit readout in bias-enhanced tunnel magneto-Seebeck effect Sci. Rep. 5 8945

Martens U, Walowski J, Schumann T, Mansurova M, Boehnke A, Huebner T, Reiss G, Thomas A and Münzenberg M 2017 Pumping laser excited spins through MgO barriers J. Phys. D: Appl. Phys. 50 144003

López-Monis C, Matos-Abiaique A and Fabian J 2014 Tunneling magnetothermopower in magnetic tunnel junctions Phys. Rev. B 89 054419

Huebner T, Martens U, Walowski J, Boehnke A, Krief J, Heiliger C, Thomas A, Reiss G, Kuschel T and Münzenberg M 2017 Enhancement of thermovoltage and tunnel magneto-Seebeck effect ratio in CoFeB based magnetic tunnel junctions by variation of the MgAl2O4 and MgO barrier thickness Phys. Rev. B 96 214435

Huebner T, Martens U, Walowski J, Münzenberg M, Thomas A, Reiss G and Kuschel T 2018 Thermal conductivity of thin insulating films determined by tunnel magneto-Seebeck effect measurements and finite-element modeling J. Phys. D: Appl. Phys. 51 224006

Mavrokefalog A, Pettes M T, Zhou F and Shia L 2007 Four-probe measurements of the in-plane thermoelectric properties of nanofilms Rev. Sci. Instrum. 78 034901

Avery A D, Purfial M R and Zink B L 2012 Observation of the planar near effect in palladium and nickel thin films with in-plane thermal gradients Phys. Rev. Lett. 109 196602

Dejene F K, Flippe J, Bauer G E W and van Wees B J 2013 Spin heat accumulation and spin-dependent temperatures in nanopillar spin valves Nat. Phys. 9 636–9

Liebing N, Serrano-Guisán S, Krzysteczko P, Rott K, Reiss G, Langer J, Ocker B and Schumacher H 2008 Biased quasiballistic spin torque magnetization reversal Phys. Rev. Lett. 101 087201
[74] Liebing N, Serrano-Guisan S, Rott K, Reiss G, Langer J, Ocker B and Schumacher H W 2012 Determination of spin-dependent Seebeck coefficients of CoFeB/MgO/CoFeB magnetic tunnel junction nanopillars J. Appl. Phys. 111 07C520

[75] Johnson M and Silsbee R H 1987 Thermodynamic analysis of interfacial transport and of the thermomagnetoelectric system Phys. Rev. B 35 4959

[76] Johnson M 2010 Spin caloritronics and the thermomagnetoelectric system Solid State Commun. 150 543

[77] Böhnert T, Dutra R, Sommer R L, Paz E, Serrano-Guisan S, Ferreira R and Freitas P P 2017 Influence of the thermal interface resistance on the thermovoltage of a magnetic tunnel junction Phys. Rev. B 95 104441

[78] Böhnert T, Serrano-Guisan S, Paz E, Lacoste B, Ferreira R and Freitas P P 2017 Magnetic tunnel junctions with integrated thermometers for magnetothermopower measurements J. Phys.: Condens. Matter 29 185303

[79] Böhnert T, Paz E, Ferreira R and Freitas P P 2018 Magnetic tunnel junction thermocouple for thermoelectric power harvesting Phys. Lett. A 382 1437

[80] Ning K et al 2017 Magneto-Seebeck effect in magnetic tunnel junctions with perpendicular anisotropy AIP Adv. 7 015035

[81] Brinkman W F, Dynes R C and Rowell J M 1970 Tunneling conductance of asymmetrical barriers J. Appl. Phys. 41 1815

[82] Meiner M, Friedrich C, Reiss G and Blügel S 2012 GW study of the half-metallic Heusler compounds Co2MnSi and Co2FeSi Phys. Rev. B 86 245115

[83] Geisler B and Kratzer P 2015 Spinocaloric properties of epitaxial Co2MnSi/MgO/Co2MnSi magnetic tunnel junctions Phys. Rev. B 92 144418

[84] Heiliger C, Zahn P and Mertig I 2006 Microscopic origin of magnetoresistance Mater. Today 9 46

[85] Amin V P, Zemen J, Železný J, Jungwirth T and Sinova J 2014 Large-tunneling anisotropic magneto-Seebeck effect in a CoPt/MgO/CoPt tunnel junction Phys. Rev. B 90 140406

[86] Miura Y, Muramoto S, Abe K and Shirai M 2012 First-principles study of tunneling magnetoresistance in Fe/MgAl2O4/Fe(001) magnetic tunnel junctions Phys. Rev. B 86 024426

[87] Tao B S et al 2015 Long-range phase coherence in double-barrier magnetic tunnel junctions with a large thick metallic quantum well Phys. Rev. Lett. 115 157204

[88] Zhang J, Zhang X G and Han X F 2012 Spinolites: Δ1 spin-filter barrier for a class of magnetic tunnel junctions Appl. Phys. Lett. 100 222401

[89] Sukegawa H, Xiù H, Ohkubo T, Furubayashi T, Niizeki T, Wang W, Kasai S, Mitani S, Inomata K and Hono K 2010 Tunnel magnetoresistance with improved bias voltage dependence in lattice-matched Fe/spinel MgAl2O4/Fe(001) junctions Appl. Phys. Lett. 96 212505

[90] Tao B, Li D, Liu H, Wei H, Feng J F, Wang S and Han X 2014 Transport properties in sputtered CoFeB/MgAl2O4/CoFeB magnetic tunnel junctions IEEE Trans. Magn. 50 4401004

[91] Scheike T, Sukegawa H, Inomata K, Ohkubo T, Hono K and Mitani S 2016 Chemical ordering and large tunnel magnetoresistance in CoFeAl/MgAl2O4/CoFeAl(001) junctions Appl. Phys. Express 9 055004

[92] Sukegawa H, Mitani S, Ohkubo T, Inomata K and Hono K 2013 Low-resistive monocristalline Mg–Al–O barrier magnetic tunnel junctions for spin-transfer magnetization switching Appl. Phys. Lett. 103 142409

[93] Heiliger C and Stiles M D 2008 Ab initio studies of the spin-transfer torque in magnetic tunnel junctions Phys. Rev. Lett. 100 186805

[94] Jaf frets H, Lacour D, Van Dau F N, Briatico J, Petroff F and Vaures A 2001 Angular dependence of the tunnel magnetoresistance in transition-metal-based junctions Phys. Rev. B 64 064427

[95] Heiliger C, Czerner M, Liebing N, Serrano-Guisan S, Rott K, Reiss G and Schumacher H W 2013 Unusual angular dependence of tunneling magneto-Seebeck effect AIP Adv. 8 115114

[96] Yang H F, Hu H K, Sievers S, Böhnert T, Tarequzzaman M, Costa J D, Ferreira R, Bieler M and Schumacher H W 2018 The magnetic tunnel junction as a temperature sensor for buried nanostructures J. Appl. Phys. 124 174501

[97] Yang H F, Hu X K, Liebing N, Böhnert T, Costa J D, Tarequzzaman M, Ferreira R, Sievers S, Bieler M and Schumacher H W 2017 Electrical measurement of absolute temperature and temperature transients in a buried nanostructure under ultrafast optical heating Appl. Phys. Lett. 110 232403

[98] Zhang J, Bachman M, Czerner M and Heiliger C 2015 Thermal transport and nonequilibrium temperature drop across a magnetic tunnel junction Phys. Rev. Lett. 115 037203

[99] Hofmeister A M 2014 Thermal diffusivity and thermal conductivity of single-crystal MgO and Al2O3 and related compounds as a function of temperature Phys. Chem. Miner. 41 361

[100] Lee S M, Cahill D G and Allen T H 1995 Thermal conductivity of sputtered oxide films Phys. Rev. B 52 253

[101] Majumdar A and Reddy P 2004 Role of electron–phonon coupling in thermal conductance of metal–nonmetal interfaces Appl. Phys. Lett. 84 4768

[102] Lin Z, Zhigilei L V and Celli V 2008 Electron–phonon coupling and electron heat capacity of metals under conditions of strong electron–phonon nonequilibrium Phys. Rev. B 77 075133

[103] Xu X, Lin W, Petit-Watelot S, Henn M, Rinnert K, Lu Y, Montaigne F, Lacour D, Andreu S and Mangin S 2016 Origins of large light induced voltage in magnetic tunnel junctions grown on semiconductor substrates J. Appl. Phys. 119 023907

[104] Huang S Y, Wang W G, Lee S F, Kwo J and Chien C L 2011 Intrinsic spin-dependent thermal transport Phys. Rev. Lett. 107 216604

[105] Meier D, Reinhardt D, Schmid M, Back C H, Schmalhorst J M, Kuschel T and Reiss G 2013 Influence of heat flow directions on Nernst effects in Py/Pt Phys. Rev. B 88 184425

[106] Schmid M, Srichandan S, Meier D, Kuschel T, Schmalhorst J-M, Vogel M, Reiss G, Strunk C and Back C H 2013 Transverse spin Seebeck versus anomalous and planar Nernst effects in permalloy thin films Phys. Rev. Lett. 111 187201

[107] www.humboldt-professor.de/en/preistraeger/ preistraeger-2014/sinova-jairo

[108] Cahill D G 2004 Analysis of heat flow in layered structures for time-domain thermoreflectance Rev. Sci. Instrum. 75 5119

[109] Jia T X, Wang S Z and Qin M H 2016 Seeking large thermoelectric effects in MgO-based tunnel junctions New J. Phys. 18 063028

[110] Daqiq R 2018 Enhancement of thermopower by structural asymmetry in double-barrier tunnel junctions J. Supercond. Nov. Magn. 31 86

[111] Flebus F, Bauer G E W, Duine R A and Tserkovnyak Y 2017 Theory of the magnon-mediated tunnel magneto-Seebeck effect Phys. Rev. B 96 094429