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Unusual angular dependence of tunneling magneto-Seebeck effect

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We find an unusual angular dependence of the tunneling magneto-Seebeck effect (TMS). The conductance shows normally a cosine-dependence with the angle between the magnetizations of the two ferromagnetic leads. In contrast, the angular dependence of the TMS depends strongly on the tunneling magneto resistance (TMR) ratio. For small TMR ratios we obtain also a cosine-dependence whereas for very large TMR ratios the angular dependence approaches a step-like function. The origin is that the cosine-dependent transmission function enters in the denominator of the definition of the Seebeck coefficient. Although the TMR and TMS are disconnected with respect to their magnitude the size of the TMR effect can be deducted from the angular dependence of the TMS effect. https://doi.org/10.1063/1.5042014

Spin caloritronics[1,2] combines the spin-dependent charge transport with energy or heat transport. Therefore, the spin degree of freedom is used in addition to thermoelectrics. The basic physics was already pointed out by Johnson and Silsbee.[3] Recently a number of new effects were discovered and discussed like spin-Seebeck effect,[4,5] magneto-Seebeck effect in metallic multilayers,[6] tunneling magneto-Seebeck effect (TMS),[7–9] thermal spin-transfer torque,[10] Seebeck spin tunneling,[11] thermally excited spin-currents,[12] magneto-Peltier cooling.[13]

In this letter we focus on the tunneling magneto-Seebeck effect (TMS)[8,9] in MgO based tunnel junctions, which we predicted in our previous paper.[7] The tunnel junction we consider consists of two ferromagnetic leads that are separated by a MgO barrier. Thereby, θ is the angle between the magnetizations of the two ferromagnetic leads. In our previous studies we found for the angular dependence of the Seebeck coefficient an almost step-like function based on *ab initio* theory[7] whereas our previous experiments show a cosine-dependence.[9] Other transport quantities like conductance or spin-transfer torque show in general a cosine or sine dependence.[14–16] Therefore, the aim of this letter is to investigate the angular dependence of the TMS on general arguments and compare these findings to *ab initio* calculations as well as experiments.

Starting point is the energy dependent transmission probability $T(E)$ from which the conductance G and Seebeck coefficient S

$$G = e^2 L_0 \qquad S = -\frac{1}{e\Theta}\frac{L_1}{L_0} \tag{1}$$

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can be calculated by the moments[17]

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

where $f(E, \mu, \Theta)$ is the Fermi occupation function at a given energy E, electrochemical potential μ, and temperature Θ. In other words, G is the area and S the center of mass of the function $T(E)(-\partial_E f(E, \mu, \Theta))$. Thus, there is no general relationship between G and S, unless for very low temperatures where a Sommerfeld expansion of the occupation function is possible. Consequently, there is also no general relationship between the TMR ratio

$$\mathrm{TMR} = \frac{G_P - G_{AP}}{G_{AP}} = \frac{R_{AP} - R_P}{R_P} \tag{3}$$

and the TMS ratio

$$\mathrm{TMS} = \frac{S_P - S_{AP}}{\min\left(|S_P|, |S_{AP}|\right)}, \tag{4}$$

where the subscript denotes parallel (P) or anti-parallel (AP) magnetic alignment of the ferromagnetic leads. This means that a high TMR ratio in a tunnel junction does not coincide with a high TMS ratio and vice versa. Moreover, limiting cases are possible as a high TMS effect for vanishing TMR effect. Figure 1 illustrates this fact by plotting the TMS ratio versus TMR ratio of different published experimental results.[8,9,18–21]

In the following, we extend our considerations to the angular dependence of the transport properties. Usually, the transmission $T(E, \theta)$ at a given energy and a relative magnetic orientation between the two ferromagnetic layers θ shows a cosine dependence going from parallel $T_P(E) = T(E, 0°)$ to anti-parallel $T_{AP}(E) = T(E, 180°)$ alignment of the ferromagnetic layers[15]

$$T(E,\theta) = \frac{T_P(E) + T_{AP}(E)}{2} + \frac{T_P(E) - T_{AP}(E)}{2}\cos(\theta). \tag{5}$$

Plugging this into Eqs. (1) and (2) leads to the cosine-like angular dependence of the conductance

$$G(\theta) = \frac{G_P + G_{AP}}{2} + \frac{G_P - G_{AP}}{2}\cos(\theta). \tag{6}$$

Consequently, the resistance $R(\theta) = 1/G(\theta)$ is not cosine-like but actually depends on the size of the TMR ratio.[14] Using the definition of the TMR ratio Eq. (3) one gets for the angular dependence of the resistance

$$R(\theta) = \frac{2R_{AP}}{\mathrm{TMR} + 2 + \mathrm{TMR}\cos(\theta)}. \tag{7}$$

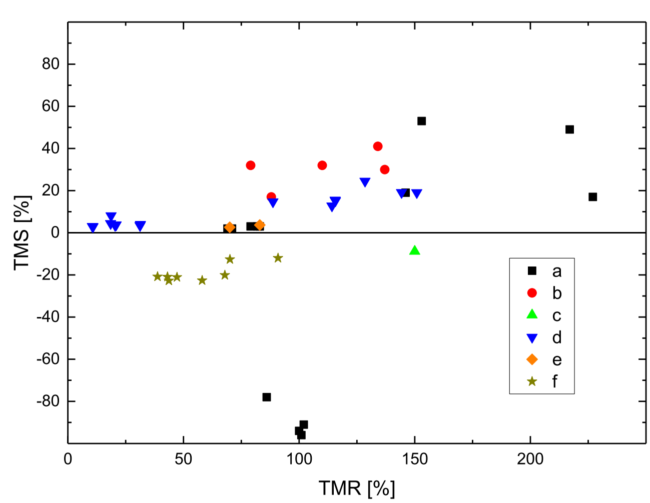

FIG. 1. TMS ratio (4) versus TMR ratio (3) for different published experimental results. Data taken from: a,[18] b,[9] c,[8] d,[19] e,[20] and f.[21]

Making a taylor expansion for small TMR ratios and in the limit of vanishing TMR ratio one reaches at

$$R(\theta) = \frac{R_P + R_{AP}}{2} + \frac{R_P - R_{AP}}{2}\cos(\theta). \tag{8}$$

This means that a cosine-like dependence for the resistance is only approximately valid if the TMR ratio is small.

Let us now turn to the TMS effect. Plugging Eq. (5) into Eqs. (1) and (2) leads to the angular dependence of the Seebeck coefficient

$$S(\theta) = \frac{S_P G_P + S_{AP} G_{AP} + (S_P G_P - S_{AP} G_{AP})\cos(\theta)}{G_P + G_{AP} + (G_P - G_{AP})\cos(\theta)}. \tag{9}$$

Using the definition of the TMR ratio in Eq. (3) we get

$$S(\theta) = \frac{S_P \mathrm{TMR} + S_P + S_{AP} + (S_P \mathrm{TMR} + S_P - S_{AP})\cos(\theta)}{\mathrm{TMR} + 2 + \mathrm{TMR}\cos(\theta)}. \tag{10}$$

Equation (10) is the main result of our paper. It shows that the angular dependence of the Seebeck coefficient is rather complex and depends on the TMR ratio. Therefore, it is worth to consider two special cases:

First, we want to consider a vanishing TMR ratio TMR = 0. Then Eq. (10) simplifies to

$$S(\theta) = \frac{S_P + S_{AP}}{2} + \frac{(S_P - S_{AP})\cos(\theta)}{2}, \tag{11}$$

which is the cosine-like angular dependence that we already obtained for the conductance in Eq. (6). Another important result is evident: although the TMR vanishes a TMS effect can exist.

Second, we want to consider very large TMR ratios. For this purpose we rewrite Eq. (10) to

$$S(\theta) = \frac{S_P + \frac{S_P + S_{AP}}{\mathrm{TMR}} + (S_P + \frac{S_P - S_{AP}}{\mathrm{TMR}})\cos(\theta)}{1 + \frac{2}{\mathrm{TMR}} + \cos(\theta)}. \tag{12}$$

In the limit of an infinite TMR ratio $S(\theta < 180°) = S_P$. On the other hand we know that $S(\theta = 180°) = S_{AP}$. Consequently, $S(\theta)$ becomes a step-like function in the limit of infinity TMR ratio

$$\lim_{\mathrm{TMR}\to\infty} S(\theta) = \begin{cases} S_P & \text{if } \theta < 180° \\ S_{AP} & \text{if } \theta = 180° \end{cases}. \tag{13}$$

Figure 2 shows the angular dependence of S for TMR ratios between these both limiting cases. This viewgraph shows that the angular dependence of S is strongly affected by the TMR ratios. Consequently, this variation with the TMR ratio can explain the different observations in the experiment

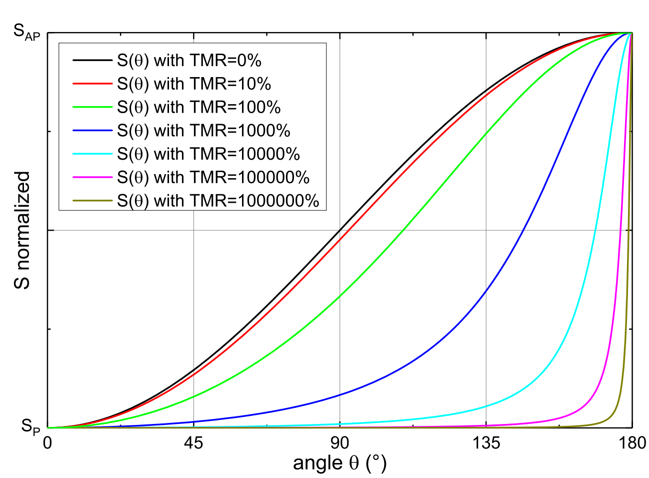

FIG. 2. Seebeck coefficient given by Eq. (10) as a function of the angle θ between the magnetic orientations of the ferromagnetic leads for different TMR ratios.

and theory for MgO based tunnel junctions because in the experiment typically TMR ratios are about 100% whereas in theory typically TMR ratios are several 1000%.

To validate Eq. (10) we compare it in Fig. 3 to our previous *ab initio* calculations.[7,8,22] The viewgraph clearly shows that Eq. (10) is indeed valid and that we have a perfect match of the *ab initio* results with the qualitative model given in Eq. (10). Consequently, our findings can explain the different observed angular dependence in theory and experiment due to different TMR ratios.

To experimentally test the predictions of Eq. (10) we compare in Fig. 4 our findings to our experimental results obtained on a nanopillar fabricated from a CoFeB/MgO/CoFeB based magnetic tunnel junction. The sample structure and setup for the electrical measurements of the TMR and thermopower have been described in Refs. 23 and 9, respectively. The nanopatterned MTJ cell comprises a 3 nm thick CoFeB free layer and is patterned into a nominally elliptical shape of 150 nm x 300 nm lateral dimensions by electron beam lithography and sputter etching. For the given device we derive a total anisotropy field $\mu_0 H_K$ of about 20 mT from measurements of the magnetization reversal asteroid as function of easy and hard axis. Note that the cell shape is likely to deviate from the nominal elliptical shape due to imperfections of the patterning process. This leads to multi domain magnetization reversal behavior especially near hard axis field orientation as indicated by multiple jumps in the magnetization loops. For the angular dependent measurements the temperature gradient was created by direct current heating with heating power up to 110 mW to generate temperature gradients of a few tens of mK across the MgO barrier. The Oersted field generated by the heater line is carefully compensated by an external field onto which the rotational field is superimposed. A 30 mT vector field is rotated in steps of 4° and the TMR and magneto thermopower are measured. Note that for the given vector field the free layer magnetization is not fully aligned to the field vector due to the influence of the effective anisotropy. The anisotropy function is determined by means of switching field angular measurements of the MTJ. The obtained critical curve of the free layer is used to calculate the magnetization direction of the free layer for each applied field vector.[24] Note that the influence of the anisotropy on the equilibrium magnetization angle leads to a sparser distribution of measurement points near the hard axis orientation and a denser distribution near the easy axis. In Fig. 4 the experimental data of TMR (top) and the Seebeck coefficient (bottom) are plotted as function of the free layer angle for comparison to theory. Note that the experimental TMS data is only plotted in the angle range where the sample shows a mainly single domain rotation of the free layer magnetization leading to missing data points near the hard axis orientation.

In particular, we compare our experimental TMR and TMS data to Eq. (7) and to Eq. (10) respectively for different TMR ratios. The experimental TMR data shows mainly a single domain

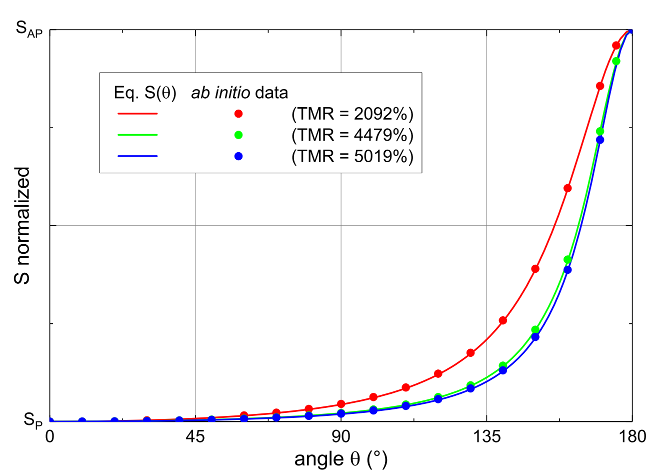

FIG. 3. Comparison of the angular dependence of the Seebeck coefficient between our previous *ab initio* calculations[7,8,22] and Eq. (10). The values are at a temperature of 300K.

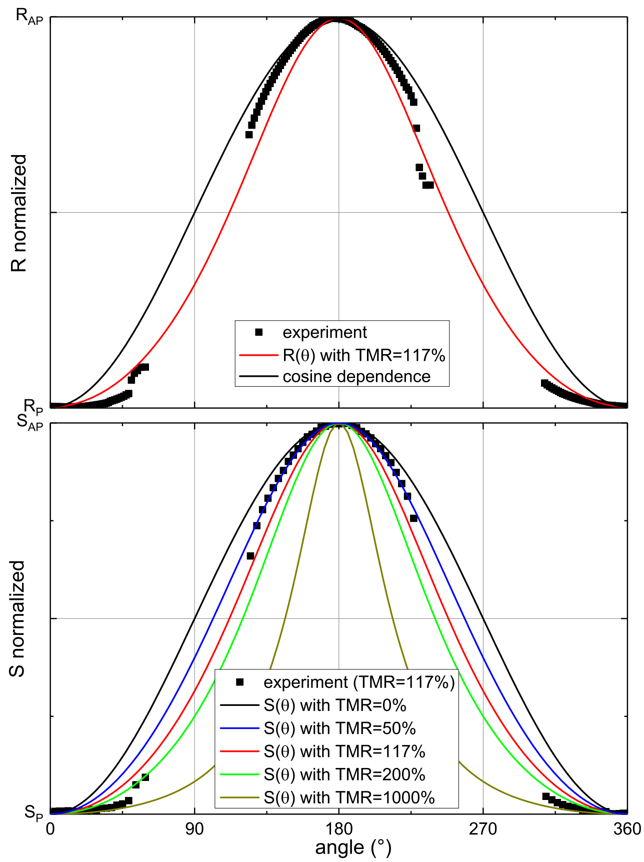

FIG. 4. Top: Comparison of the angular dependence of the resistance between our experimental data and Eq. (7). In addition, we plot a fictitious cosine dependence. Bottom: Comparison of the angular dependence of the Seebeck coefficient between our experimental data and Eq. (10) for different TMR ratios. The TMR ratio in the experiment is 117%.

behavior and thus a smooth angular dependence with a TMR ratio of about 117 % (see Fig. 4 top). However, near 60° and 240° of the free layer magnetization steps of the TMR curves indicate the influence of small non-uniform domains near the switching across the hard axis. As a consequence of these steps a variety of theoretical curves with TMR ratios around 100 % would fit the experimental angular dependence of TMR and TMS. Thus, fitting the experimental TMS data to Eq. (10) a TMR ratio of 78 % is derived, which is close to the measured TMR ratio in Fig. 4 top keeping in mind the scattering of data points. Note that around $\Theta = 180°$ the experimental data shows a better agreement with the theoretical curve for a lower TMR of 50 % whereas around 0° it is in better agreement with the predictions for higher TMR ratios. This could be related to imperfections of the patterning process resulting in a complex sample anisotropy. Such a deviation of the determined anisotropy function by the astroid method[24] from the real, rather complex, effective anisotropy causes an error in the calculated free layer magnetization orientation and thus in the dependence of TMR and TMS curves. Nevertheless, there is a significant deviation from the cosine-like dependence in both TMR and TMS curves, as a consequence of the large measured TMR ratio, as pointed out previously in the text.

This is also an example how the size of the TMR ratio can be extracted from the qualitative angular dependence of the TMS. On a first view this is somehow surprising, because due to the definition given in Eq. (1) there is no general relationship between the conductance and the Seebeck coefficient, unless for very low temperatures where a Sommerfeld expansion of the occupation function is possible. In the present work Eq. (10) was exemplarily compared to experimental data of one device with the given TMR of 117 %. In future studies the angular TMS dependence should be scrutinized over a range of TMR values to validate the predictions of Eq. (10). Preferably these experiments should be carried out on micrometer scale circular devices with low shape anisotropy to ensure a controlled single domain rotation over the complete angle range.

In summary, we derived an expression (10) for the angular dependence of the Seebeck coefficient. It turns out that the angular dependence is complex and strongly dependent on the TMR ratio. The angular dependence varies between a cosine-like dependence for a vanishing TMR ratio and a step-like function in the limit of an infinity TMR ratio. This variation explains the different experimental and theoretical observation of the angular dependence, because in theory the TMR ratio is at least one order of magnitude larger than in experiment. On the other hand it is also possible to deduct the size of the TMR ratio by analyzing the qualitative angular dependence of the TMS. Further, we showed that a TMS effect can exist even when no TMR effect is present.

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