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Evidence for strong magnon contribution to the TMR temperature dependence in MgO based tunnel junctions

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We have prepared MgO based magnetic tunnel junctions which show up to 143% tunneling magnetoresistance (TMR) ratio at room temperature and 205% at 12 K. This TMR temperature dependence is mainly caused by a strong temperature dependence in the antiparallel magnetic state, while in the parallel state the change of conductance is small. We found that a modified version of the magnon excitation model may be applied to these MgO magnetic tunnel junctions. If the thermal smearing of the tunneling electron's energy is included it is possible to fit the temperature dependence. We will show the results for our data and we have also tested our model successfully on data from other publications.

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I. INTRODUCTION

Since the discovery of the tunneling magnetoresistance (TMR) effect[1] its amplitude has strongly increased. While early experiments showed significant TMR only at low temperature,[2] the effect was later also shown at room temperature,[3,4] leading to ratios as high as 472% (Ref. 5) to date. Yet, the TMR still increases significantly if the junctions are cooled to low temperatures.[6]

While the changing TMR ratio in magnetic tunnel junctions (MTJs) with alumina barriers goes along with comparable conductance changes in both magnetic states,[7,8] this is not the case in newer junctions with MgO barriers and high TMR ratios.[5,9,10] Even in systems with very different electrodes[11] the decrease of TMR with rising temperature is mostly carried by a change in the antiparallel conductance. The parallel conductance changes so little that it seems roughly constant, if compared to the antiparallel conductance.

Different models are at hand for the mechanism of this temperature dependence. One is the model by Shang and Moodera[8,12] which is based on Jullière's model with a directly temperature dependent spin polarization of the ferromagnetic electrodes.

The other model by Zhang *et al.*[7] is based on two-dimensional spin waves excited by tunneling electrons at the insulator ferromagnet interface. In this model a lower energy cutoff E_c was introduced to get a finite number of excited magnons at a nonzero temperature. The physical representation of this cutoff can be, e.g., a maximum coherence length in the magnetic structure or an anisotropy for the spins present at the interfaces.

For incoherent tunneling this model gives a TMR-voltage-dependence at zero temperature $T=0$ and low bias V of

$$TMR(0,V) = TMR(0,0) - Q\frac{SeV}{E_m}\frac{R_{AP}(0,0)}{R_P(0,0)}\left(\frac{1}{\xi} - \xi\right), \qquad (1)$$

where $TMR(0,0)$, $R_P(0,0)$, and $R_{AP}(0,0)$ are the TMR ratio and the resistance in the parallel and antiparallel state, with $TMR=(R_{AP}-R_P)/R_P$. The parameter Q describes the probability of a magnon to be involved in the tunneling process and will be used as a fitting parameter. S is the spin parameter, while E_m is related to the Curie temperature $E_m = 3k_BT_C/(S+1)$ of the ferromagnetic electrodes. It should be noted that in both Eq. (1) and the following temperature dependence Q is scaled by S/E_m. Therefore their actual values do not change the temperature dependence but only the numerical value of Q. The parameter ξ is the ratio of the products of density of states in parallel and antiparallel configuration: $\xi=2\rho_M\rho_m/(\rho_M^2+\rho_m^2)$. In our case this is the same as the ratio of current or resistance in both states $\xi = j_{AP}(0,0)/j_P(0,0)=R_P(0,0)/R_AP(0,0)$.

Then the temperature dependence of the resistance in parallel $R_P(T,V=0)$ and antiparallel $R_{AP}(T,V=0)$ state at zero bias V can be expressed as

$$R_P(T,0) = R_P(0,0)\left[1 + Q\xi\frac{2S}{E_m}k_BT\ln\left(\frac{k_BT}{E_c}\right)\right]^{-1}, \qquad (2)$$

$$R_{AP}(T,0) = R_{AP}(0,0)\left[1 + Q\frac{1}{\xi}\frac{2S}{E_m}k_BT\ln\left(\frac{k_BT}{E_c}\right)\right]^{-1}. \qquad (3)$$

Here, E_c is the magnon energy cutoff energy. Further details can be found elsewhere.[7,13]

Until now another fundamental intrinsic mechanism has been disregarded as very small: In a free electron, incoherent tunneling model the thermal smearing of the electron energies decreases the effective barrier height with increasing temperature.[14] This effect could be ignored when the changes in conductivity were substantially higher due to other (extrinsic) effects; but this is not the case for newer systems with higher TMR, especially in the parallel magnetic state where the overall change in conductance is very small. Also for coherent tunneling a change of the conductance is expected because additional conductance channels above and below the Fermi energy E_F can be opened.

No theoretical description of coherent tunneling including thermal smearing has been done so far. In this paper we will show that the extension of the magnon-assisted tunneling model by thermal smearing can also be successfully applied as a phenomenological model to MgO based MTJs.

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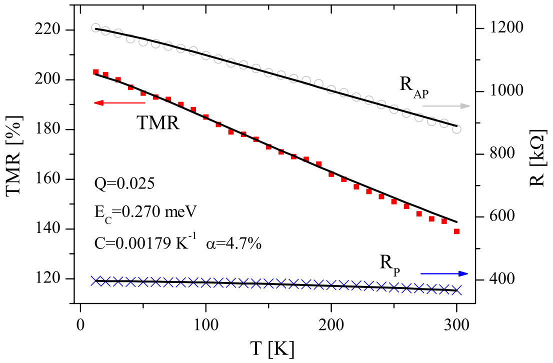


FIG. 1. (Color online) Resulting fit for the TMR temperature dependence of our MgO MTJ using the magnon excitation model and thermal smearing.

II. PREPARATION

The magnetic tunnel junctions are prepared in a magnetron sputter system with a base pressure of 1×10^{-7} mbar. The layer stack is Ta 10/Cu 30/Ta 10/Cu 5/$Mn_{83}Ir_{17}$ 10/$Co_{40}Fe_{40}B_{20}$ 2.5/MgO 1.5/$Co_{40}Fe_{40}B_{20}$ 4/Ta 10/Cu 30/Ru 10 (all values in nm) on top of a thermally oxidized (50 nm) silicon (100) wafer. To activate the exchange biasing and for the crystallization of the MgO barrier, the layer stack is annealed after sputtering at 623 K for 60 min in a magnetic field of 6500 Oe. The stack is patterned by *e*-beam lithography and ion beam etching. The resulting patterns are ellipses with an aspect ratio of 3 and long axes of 6, 1.5, and 0.75 μm. These structures are capped with gold pads.

All measurements are done by a conventional two probe technique in a closed cycle helium cryostat (Oxford Cryodrive 1.5) with a temperature range of 12–330 K. We have also performed inelastic electron tunneling spectroscopy (IETS) measurements at 12 K utilizing a lock-in technique with a bias modulation of 2 mV at 500 Hz.

III. RESULTS AND DISCUSSION

The measurement of a typical junction's resistance is shown in Fig. 1. The element shows a TMR of 143% at RT, increasing to 205% if cooled to 12 K. This is a relative increase of 43%, while at the same time the junctions resistance changes 36% and 8% in the antiparallel and parallel state, respectively.

Compared to our CeFeB/Al-O/CeFeB junctions[15] this change in resistance in the parallel case is smaller. This suggests that also smaller effects like thermal smearing become more important here.

The thermally induced change of the resistance is different for alumina and MgO based MTJs, but IETS shows very similar properties. IET spectra of a MgO MTJ in a parallel and antiparallel state are shown in Fig. 2 whereas spectra of our alumina junctions can be found elsewhere.[16]

The zero bias anomaly which is related to magnons[13,16] and the typical phonon peaks—at around ±81 mV for the Mg-O (Ref. 17) and ±120 mV for the Al-O phonon—can easily be identified. This gives strong evidence that magnons are also involved in the tunneling process for MgO. Moreover, the spectra look similar, both lacking substantial features hinting to a macroscopic effect of coherent tunneling.

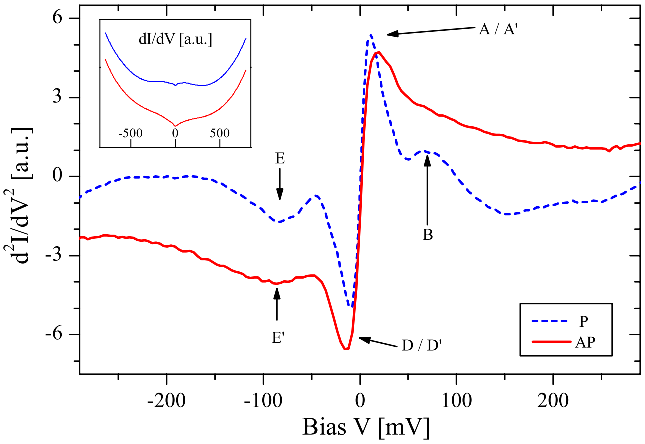


FIG. 2. (Color online) IET spectra of a MgO MTJ in a parallel (dashed line) and antiparallel (solid line) state at 12 K. Typical magnon (A, D) and phonon (B, E) peaks can be identified in parallel and antiparallel (e.g., A′) configuration.

A. First model: No convergence for high TMR

The basic assumption of the model by Shang[8] is a temperature dependence of the ferromagnets spin polarization which is proportional to the magnetization. Using Julliere's formula leads to a resistance in antiparallel state which strongly decreases with rising temperature. In the parallel case the resistance is rising with temperature. As the opposite behavior—a small decrease of the resistance in parallel state with rising temperature—is found in many magnetic tunnel junctions[5,8,9,18–20] an additional term has to be introduced. This term must be spin-independent and have a temperature dependence which shows a strong decrease in resistance in order to compensate the basic $dR_P/dT>0$ behavior. Shang *et al.* proposed that this could be hopping through localized states[8] because it can fit the observed power law with an exponent of about 4/3.

However, this model cannot hold for junctions with higher TMR. If one investigates junctions with higher TMR the model describes a stronger temperature dependence of the resistance in both magnetic states. Therefore the spin-independent term in this model must also increase in order to compensate the stronger basic $dR/dT>0$ tendency. One would assume that a higher TMR is linked to a MTJ of better quality with regard to barrier structure and magnetism. A spin independent term has the tendency of lowering the TMR and is a sign for a barrier of inferior quality. Then it would be very unlikely that a spin independent term can be higher in a MTJ with higher TMR. Yet, it is unlikely but not impossible.

What counts more is that in every single case the two independent contributions to the tunneling current would have to be "fine tuned" to exactly cancel each other out to give a small dependence with $dR_P/dT<0$. For a given set of samples this could be possible by incident, but it would be physically unreasonable to expect this in general for all MTJs. To the best of our knowledge there is not a single publication which states a tendency of R_P to rise with rising temperature. On the contrary the size of the dependence is nearly the same in all publications. The chance that the spin-independent and spin-dependent term cancel each other out in every sample prepared by different groups and different methods and with different materials is too small to be a reasonable explanation. Therefore we think this model is not able to give a physical explanation of the T-dependence in high TMR junctions.

B. Magnon model expanded by thermal smearing

In the magnon model, a surface magnon can be emitted or absorbed by a tunneling electron, opening additional conductance channels. Because of angular momentum conversation the spin of this electron has to be flipped and it contributes to another spin-channel. Therefore the overall conductance is always a mixed state of both parallel and antiparallel state if magnons are excited. If only incoherent tunneling is considered the electronic band structure of the ferromagnetic electrodes can be simply described as the density of spin-up and spin-down states. Moreover, if only the states at the Fermi energy are taken into account a spin polarization of the electrodes can be defined.

The most notable result is the simultaneous modeling of the low temperature dependence of the conductivity in the parallel state and the large dependence in the antiparallel state for high TMR junctions without introduction of additional contributions to the conductance.

In principle, the proposed magnon assisted tunneling model by Zhang *et al.*[7] is also able to theoretically describe coherent effects. The barrier Hamiltonian H_B is a function of the annihilation operators for electrons and magnons as well as the transition matrix. The latter depends on the wave vectors $\mathbf{k}$ and $\mathbf{k}'$ of the initial and final state. In this situation coherent tunneling can mathematically be described where $\mathbf{k}=\mathbf{k}'$ or $\mathbf{k}=\mathbf{k}'\pm\mathbf{q}$ if magnons are involved; but the Hamiltonian in this form is not applicable to any experimental data. The full band structure and all the energy- and wave-vector-dependent transmission matrix elements would have to be calculated to get an exact description.

In the former case of incoherent tunneling two simplifications were made, namely the introduction of an effective spin polarization and the nonenergy dependent transmission matrix. In our case, two simplifications have also to be made to do quantitative analysis of the presented measurements.

(i) The spin polarization P in alumina based MTJs is often interpreted as the difference of the itinerant spin-up and spin-down electrons at the Fermi energy. This is certainly incorrect for our case. Here, the parameter P specifies the difference between the number of spin-up and spin-down electrons tunneling from one ferromagnet through the barrier into the other ferromagnet and is an effective value averaged over the total tunneling current.

(ii) The probability for electrons of different energy and spin tunneling from their initial to their final state (i.e., the transmission matrix elements) is also taken as an effective, averaged value. We think that these effective values can describe real MTJs that show no sign of sharp features indicating coherent tunneling in, e.g., IETS curves, as shown before.

In addition to the basic model by Zhang *et al.* the intrinsic effect of thermal smearing has to be considered. With the above-mentioned assumptions an increasing temperature and the accordingly wider Fermi edge leads to a smaller effective barrier height not only for alumina but also for MgO based MTJs and, therefore, an additional increase in conductance. For nonmagnetic Al/Al-O/Al tunnel junctions this change is only a few percent depending on the barrier properties.[14] Thus it could be neglected for alumina MTJs with strong temperature dependence caused by other (extrinsic) effects. However, this is not true for MTJs with higher TMR ratios, as the conductance change in the parallel state becomes very small.

We will see shortly that the magnon model alone cannot be used to fit the temperature dependence in both magnetic states correctly. Adding the thermal smearing can improve the fit quality and give a self-contained explanation for the characteristics of the temperature dependence.

We can estimate the influence of the thermal smearing using

$$\frac{G(T)}{G(0)}=\frac{CT}{\sin(CT)}, \tag{4}$$

with $C=1.387\times10^{-4}d/\sqrt{\phi}$ where d is the barrier thickness (in Å) and ϕ the barrier height (in eV).[14]

For our junction we have a thickness of $d=1.5$ nm. Using a barrier height of $\phi=3.5$ eV, which is half of the MgO band gap,[21] results in $C=1.222\times10^{-3}$. To get a better idea of the size of the thermal smearing contribution we define $\alpha=1-\frac{\sin(C\times300\text{ K})}{C\times300\text{ K}}$, thus the change in resistance from 0 to 300 K. The value for C then corresponds to $\alpha=1.8\%$. This is in the same order as the overall temperature dependence in the parallel case and should not be neglected.

As a first order approximation we can, therefore, multiply the additional term from Eq. (4) and use C as an additional fitting parameter:

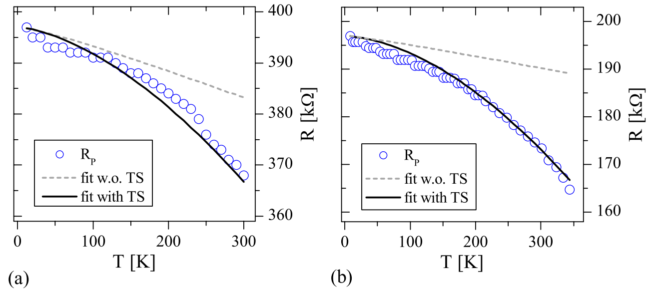
$$R_\gamma(T,0)=R_\gamma(0,0)\frac{\sin(CT)}{CT}\left[1+Q\beta_\gamma\ln\left(\frac{k_BT}{E_c}\right)\right]^{-1}. \tag{5}$$

Here $\gamma=(P,AP)$ denotes parallel and antiparallel state, respectively, with $\beta_P=Sk_BT\xi/E_m$ and $\beta_{AP}=Sk_BT/(\xi E_m)$.

The first step to apply this model is to get the parameter Q from the TMR(V)-curve at 0 K using Eq. (1). We approximate this with our measurements at 12 K. The MTJs parameters are $R_P(0,0)=397$ kΩ, $R_{AP}(0,0)=1203$ kΩ, $TMR(0,0)=205\%$, and $\xi=3.279$. The other parameters used are $S=3/2$, $E_m=121$ meV. The fit results in $Q=0.0242$.

With these values, we can fit the overall temperature dependence with Eq. (5) and get $E_c=0.270$ meV and $C=1.79\times10^{-3}$ K^{-1} or $\alpha=4.7\%$. The fit shows very good agreement with the measured data and is shown in Fig. 1. The size of the thermal smearing is also in good agreement with the theoretical expectation.

A comparison between the pure magnon model and our enhanced model is shown in Fig. 3. Clearly, the simple magnon model underestimates the temperature dependence in the



(a) (b)

FIG. 3. (Color online) Fit results for the magnon excitation model without and with the thermal smearing extension for (a) our data and (b) the data of Parkin *et al.* (Ref. 9). (While in both cases R_P and R_{AP} are fitted, only the resistance in parallel state is shown—the improvement is largest here.)

parallel state. As the overall change of the resistance with temperature is very small in high TMR junctions, the change due to other small (nonmagnon) effects cannot be neglected here. Not only the improvement to the fit provided by our model but also the good agreement of theoretical expectations and the gained fit value for the thermal smearing suggest that thermal smearing is a reasonable explanation. Furthermore, both magnon-excitation and thermal smearing are intrinsic effects which are present in every magnetic tunnel junction. Together a simple self-consistent explanation for the temperature dependence in high TMR MTJs can be provided.

IV. OTHER DATA

We applied this model to other data available. First, the work by Parkin *et al.*[9] was investigated.[22] As the barrier used has a much higher thickness of 2.9 nm we would expect a stronger temperature dependence of the thermal smearing according to Eq. (4). The calculation gives an α of 7% (ϕ=3.5 eV).

As we have no TMR(V)-data available we assume that the parameter Q is the same as in our junctions. For a general test of our model this seems adequate due to the similar layer stack. The result of the fit is shown in Fig. 4. The cutoff energy of E_C=0.116 meV corresponds to 1.35 K and is in the same range as for our junctions. The thermal smearing has a constant of C=0.002 56 K^{-1} or α=9.5% in very good agreement with the expectation. The parallel conductance change is again almost entirely caused by thermal smearing which is, therefore, even less ignorable.

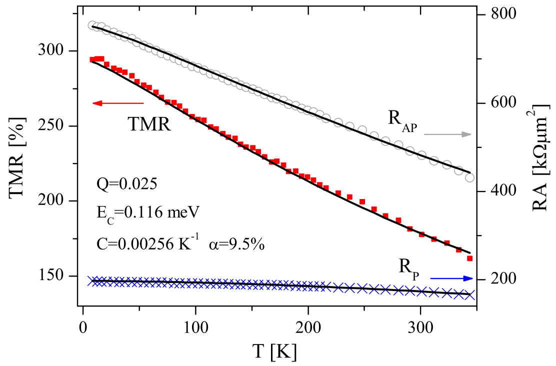


FIG. 4. (Color online) Resulting fit using our model including thermal smearing on the data of Parkin *et al.* (Ref. 9).

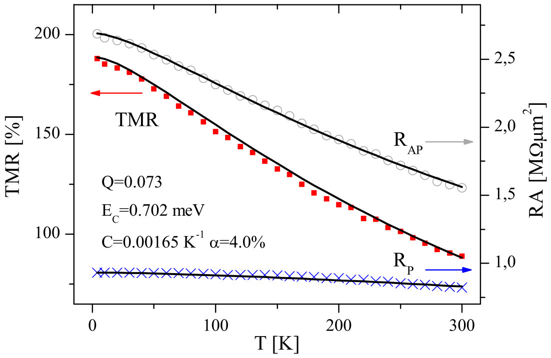


FIG. 5. (Color online) TMR temperature dependence of junctions by Ishikawa *et al.* (Ref. 11) fitted using our enhanced model.

We also tested our model on data by Ishikawa *et al.*[11] They used a tunnel junction with two different electrodes, one is a Heusler alloy and the other one CoFe resulting in a TMR of 90% at room temperature. While it is likely that the parameters Q and E_c are different for these different magnetic materials, the amount of thermal smearing should not depend on the magnetic properties. A barrier thickness of 2.4 nm and a height of 3.5 eV lead to α=4.9%.

Again the data for Q is not available. As the magnon spectrum is expected to be different for a Heusler alloy compared to Co-Fe-B we use Q as another free parameter. The result of the fit can be seen in Fig. 5. Here, the fit reproduces the change from a concave to a convex shape of the TMR curve. The parameters are Q=0.073, E_C=0.702 meV corresponding to 8.1 K and C=0.001 65 K^{-1} or α=4.0%. This is again in good agreement with the expected value. While the thermal smearing is nearly the same size as in our junction, the other parameters differ stronger, which is an expression of the overall stronger temperature dependence. Please note that also for $\mathrm{Co_2MnSi/Al\text{-}O/CoFe}$ a considerably stronger bias voltage dependence and accordingly higher Q has been found.[15]

V. SUMMARY

We have prepared MgO based magnetic tunnel junctions which show up to 143% TMR at room temperature and 205% TMR at 12 K. This TMR temperature dependence is mainly based on the strong temperature dependence in the antiparallel magnetic state, while the change of conductance is only small in the parallel state. This is the case for all MTJs with high TMR we investigated and can basically be understood by the model of magnon assisted tunneling.

For quantitative agreement with the experiment, however, it is not sufficient. Additionally taking the thermal smearing into account in a phenomenological model, we obtained a very good agreement of model and experimental data. Thus this effect cannot be neglected for high TMR junctions because of the very small overall change of conductance in the parallel state. Our effective, averaged values seem justified, as there is no evidence for macroscopic coherent effects, e.g., in IET spectra. We have also tested our model on data from other groups for magnetic tunnel junctions with high TMR ratios. The change in the fit parameters can be attributed to the differences in the junctions used but the agreement of the fits with the experimental data remains very good.

We suggest that the tailoring of the magnon spectrum is crucial for getting less temperature dependence and, therefore, a higher TMR ratio at room temperature.

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