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Barrier thickness dependence of the magnetoresistance in TaO_x magnetic tunnel junctions

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A systematic study has been conducted on the dependence of the magnetoresistance (MR) ratio on the barrier thickness in TaO_x-based magnetic tunnel junctions. The relatively low MR ratio (<10%) for the thinnest barriers studied decreases rapidly with increasing thickness. From Rutherford backscattering analysis and current-voltage measurements evidence for a Ta thickness dependent oxidation rate has been found. Photoconductance spectra measured on the same junctions indicate changes in the hot-electron transport *into* the barrier, independent of the barrier asymmetry. These changes further indicate a modification of the junction structure with increasing Ta thickness, which can have a strong adverse effect on the spin polarization of the tunnel current. © 2005 American Institute of Physics. [DOI: 10.1063/1.1872199]

I. INTRODUCTION

Magnetic tunnel junctions (MTJs) are currently of high interest as sensor elements in the field of hard disk recording and as solid state memories, due to their high magnetoresistance (MR) ratios. MTJs consist of a thin insulating tunnel barrier, sandwiched between two magnetic electrodes. For the implementation in nonvolatile magnetic memories (MRAMs), downscaling of the junction area becomes an important issue.¹ To maintain or increase the readout speed, it is essential that the junction resistance does not increase, which would otherwise lead to an unacceptable electronic noise level. The resistance-area product ($R \times A$) is therefore the parameter which has to be reduced. Since the tunneling probability depends exponentially both on the barrier thickness as well as on the (square root of the) barrier height,² a reduction of the $R \times A$ product can therefore be accomplished by reducing either of the two.

The material which is most often used as the tunnel barrier is aluminum oxide. By oxidizing a thin Al layer the highest MR ratios at room temperature, up to ~70%, have been achieved.³ However, experimentally it has been found that reducing the $R \times A$ product by reducing the thickness of the deposited Al layer only works down to a certain limiting thickness, corresponding to a $R \times A$ product of ~200 $\Omega \mu\text{m}^2$. Oxidizing even thinner Al layers results in a degradation of the junction properties, leading in particular to a reduced MR ratio.⁴ A different method for reducing the $R \times A$ product would be by choosing a barrier material with a lower barrier height as compared to AlO_x.

From previous work barrier heights for plasma-oxidized AlO_x junctions have been determined with a photoconductance technique, revealing that the barrier height in such junctions is typically 2.5–3 eV.⁵ By using a barrier material

with a much lower bandgap, a reduced barrier height might be expected. One possible candidate material is TaO_x, which has a bandgap of ~4.2 eV (for bulk Ta₂O₅),⁶ compared to ~8 eV for Al₂O₃.⁷ Barrier heights obtained from fits of the Simmons² or Brinkman⁸ models to the current-voltage characteristics indeed showed significantly lower barrier heights,^{9,10} viz. in between 0.3 and 1 eV. However, only small MR ratios (<10%) have been observed. At present, it is not known why the MR ratio observed in these studies, was limited to such low values.

These low MR ratios might be of an intrinsic origin, i.e., a lower effective spin polarization at the TaO_x/Co₉₀Fe₁₀ interface, as compared to the AlO_x/Co₉₀Fe₁₀ interface. This would then be caused by different relative contributions of *s* and *d*-type electron states (with different polarizations) to the tunnel current, as suggested by Sharma *et al.*¹¹ Alternatively it might be of extrinsic origin, related to a lower structural quality of the tunnel barrier, e.g., caused by defects or impurities.^{12,13} It is difficult to extract information on the tunneling spin polarization itself in these heavy metal oxide barriers, as shown by Kant *et al.*¹⁴ Instead, we will focus on the structural origin of the electron transport characteristics.

In this paper we present results from a systematic study of the dependence of the MR ratio on the thickness of TaO_x barrier layers. A strong decrease of the MR ratio with increasing Ta thickness has been found. From structural and *I(V)* measurements evidence for nonhomogeneous oxygen growth has been found. Photoconductance experiments show distinct changes of the net hot-electron transport from the two electrodes with increasing thickness. Although an intrinsic decline of the spin-polarization cannot be ruled out, our results indicate a change of the structural quality of the tunnel junction with increasing thickness. This may already severely deteriorate the spin polarized tunnel process, leading to a lower MR ratio.

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II. EXPERIMENTAL

All junctions studied in this work have been made by sputter deposition through metal contact masks onto glass substrates. The following layer stack has been used: Glass/3.5 Ta/3.0 Ni₈₀Fe₂₀/10.0 Ir₂₀Mn₈₀/2.5 Ni₈₀Fe₂₀/1.5 Co₉₀Fe₁₀/TaO_x barrier/4.0 Co₉₀Fe₁₀/10.0 Ni₈₀Fe₂₀/3.5 Ta, with all thicknesses in nanometers. The square junction area is varied between 60×60, 110×110, and 200×200 μm². The TaO_x tunnel barrier is formed by first depositing a thin Ta layer over the complete substrate followed by oxidation in an oxygen plasma. The thickness of the Ta layer is varied between 0.8 and 1.4 nm, and the oxidation time between 5 and 240 s. No annealing steps were carried out afterwards. In this way exchange-biased MTJs with MR ratios of up to 7% at room temperature could be realized.

For the conductance, magnetoconductance, and photo-conductance measurements, all carried out at room-temperature, a home-built setup has been used.¹⁵ All measurements were carried out in a four point configuration. To study the oxygen content in the tunnel barrier, Rutherford backscattering spectrometry (RBS) experiments have been carried out on selected planar layer stacks, on Si substrates. The energy spectrum of 1.4 MeV He⁺ ions, backscattered from the sample under an angle of ~80°, is studied.

III. TRANSPORT AND STRUCTURAL STUDIES

In Fig. 1(a) the MR ratios of TaO_x MTJs, for various Ta thicknesses and oxidation times, are shown. For a given Ta thickness, the MR ratio shows a maximum as a function of oxidation time. For AlO_x MTJs this behavior is well-known, and is attributed to the complete oxidation of the barrier layer, without oxidizing the underlying magnetic electrode.^{16,17} The observed magnitude of the MR ratio is comparable to previously determined values.^{9,10} The interesting feature to be seen in Fig. 1(a) is that the maximum attainable MR ratio drops sharply with increasing Ta thickness, and completely vanishes for a thickness of 1.4 nm or more. In contrast, for junctions based on AlO_x barriers, the maximum MR ratio is mostly independent of the Al thickness. Only for extremely thin Al layer thicknesses, below roughly 0.6 nm, a reduction of the MR ratio is observed, due to the presence of pinholes.¹⁸ As discussed before, the low MR ratio as well as its strong thickness dependence might both be of intrinsic or extrinsic origin. In this section we will focus on the latter, by a study of the structural properties.

In Fig. 1(b) the $R \times A$ product is given for all deposited TaO_x junctions. For each Ta thickness, a monotonic increase with oxidation time is observed, similar to that commonly observed for AlO_x.^{5,19} This is related to the fact that for longer oxidation times, more and more oxygen is incorporated in the barrier, improving the insulating properties of the junction. However, for the same oxidation time the $R \times A$ product *decreases* with increasing Ta thickness. If a simple oxidation front would move through the Ta layer, the $R \times A$ product would be independent of the Ta layer thickness. This picture is apparently incorrect. To clarify the observed behavior, RBS analysis was carried out on similar planar layer stacks.

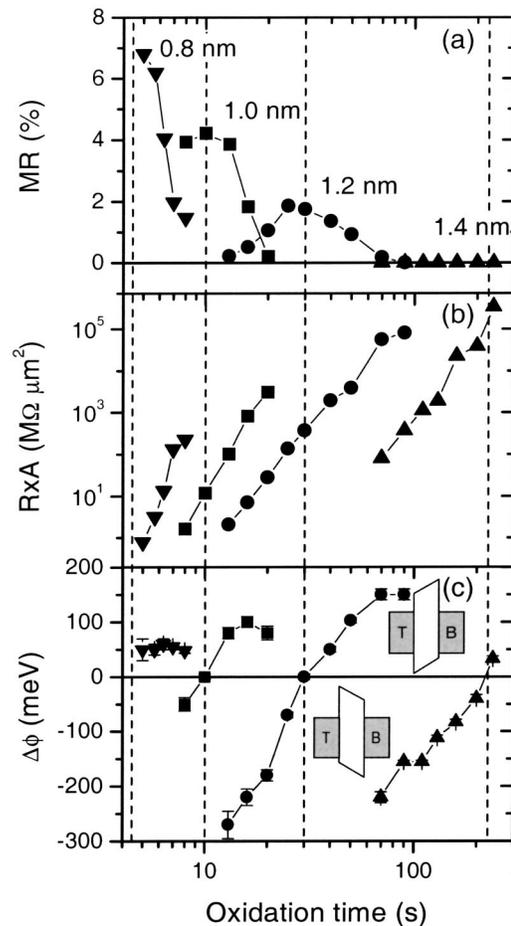


FIG. 1. For MTJs with TaO_x based tunnel barriers, (a) the MR ratio, (b) the $R \times A$ product, and (c) the barrier asymmetry $\Delta\phi$ as determined by photo-conductance are shown. In (a) the thicknesses of the deposited Ta layers are shown. The vertical dashed lines correspond to the oxidation times where the barrier is symmetric and the MR ratio has a maximum. The insets in (c) represent energy diagrams, defining the sign of the barrier asymmetry relative to the top T and bottom B electrode.

In Figs. 2(a) and 2(b) the determined oxygen content from RBS is given as a function of oxidation time (for a constant Ta thickness), as well as as a function of Ta thickness (for a constant oxidation time), respectively. As shown

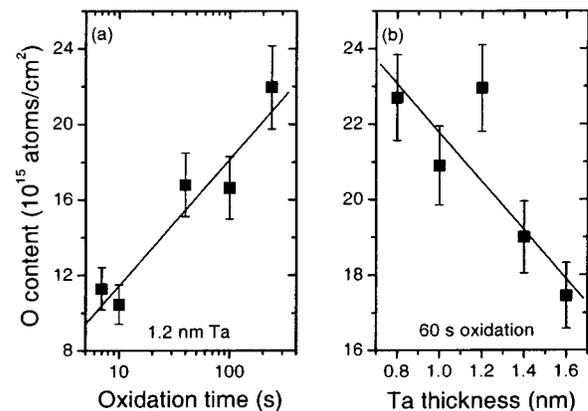


FIG. 2. Oxygen content in TaO_x MTJs as determined by RBS, for (a) constant Ta thickness and varying oxidation time, and (b) constant oxidation time and varying Ta thickness.

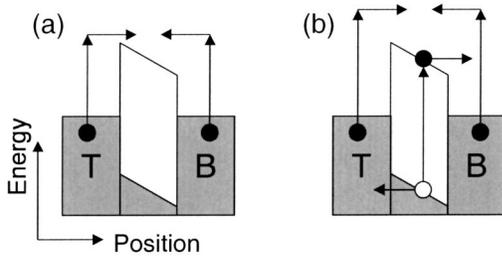


FIG. 3. Schematic energy diagrams illustrating the two separate contributions to the photocurrent. (a) For low photon energies only excitations from the two electrodes contribute. (b) For photon energies above the insulator bandgap additional excitations in the barrier itself contribute to the net photocurrent. T and B represent the top and bottom electrode, respectively.

in Fig. 2(a), the incorporation of oxygen follows a simple $\log(t)$ behavior, similar to what is found for AlO_x .¹⁹ However, with increasing Ta thickness, a significant reduction in the oxygen content, after a fixed oxidation time (60 s), is observed [Fig. 2(b)], consistent with the observed decrease of the $R \times A$ product. This directly contradicts the naive picture of a simple oxidation process where the time-dependent oxidation rate is independent of the original Ta layer thickness. For AlO_x it has been argued that, after the first few monolayers of “instantaneous” oxidation, the oxidation proceeds predominantly along metal grain boundaries.^{19,20} For an increase in grain sizes with increasing layer thickness, the oxidation rate after a given oxidation time is then expected to decrease with increasing Ta layer thickness. Furthermore, since Ta expands almost 250% after oxidation, compared to only 25% for Al,¹⁰ this will lead to a less uniform barrier layer. These two effects would indeed lead to a decrease of the $R \times A$ product with increasing Ta layer thickness, after a fixed oxidation time. Also, an increase of the barrier inhomogeneity with increasing Ta barrier thickness might explain the observed lower value of the MR ratio. It is then assumed that the effective spin-polarization is negatively affected by an increasing probability of adverse conduction processes via nonmagnetic barrier or interface states.²¹ Additionally, s - d interfacial scattering, caused by increased roughness or impurities can substantially reduce the MR ratio.¹³

IV. PHOTOCONDUCTANCE

To further investigate the influence of the barrier thickness on the electron transport, photoconductance experiments have been conducted. As discussed in a previous paper,²² the photocurrent across a low bandgap insulator such as TaO_x consists of two separate contributions. For photon energies, $h\nu$, below the bandgap only excitation in the electrodes contribute to the photocurrent [Fig. 3(a)]. For energies above the bandgap, electron-hole pairs can be excited in the barrier itself, which gives rise to an additional photocurrent contribution [Fig. 3(b)]. From the voltage dependence of the latter process, the sign and magnitude of the barrier asymmetry, $\Delta\phi$, can be extracted, as shown in Fig. 1(c).

In Fig. 4 representative photoconductance spectra are shown for both illumination of the top and bottom electrode. A positive photocurrent corresponds here to an electron flow

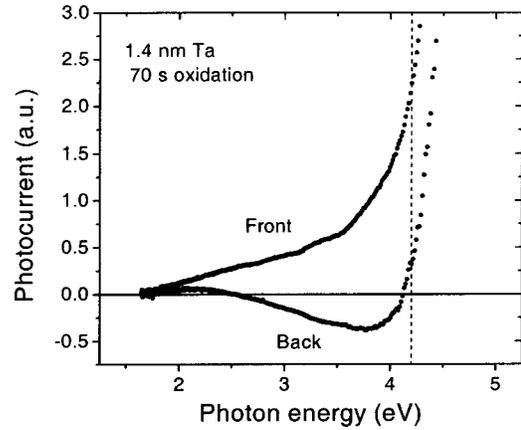


FIG. 4. Photocurrent vs energy curves for light incident on the top (front) and bottom (back) electrode, for a typical TaO_x MTJ. The vertical dashed line indicates the onset of interband excitations in the barrier layer. A positive photocurrent corresponds to an electron flow from top to bottom electrode. No bias voltage is applied.

from top to bottom electrode. One can clearly see that the contributions from excitations in the electrodes ($h\nu < 4.2$ eV) strongly depend on the direction of illumination. A clear transition from positive to negative photocurrents is visible when switching from front to back illumination. Since the bandgap of TaO_x is only 4.2 eV,⁶ and since the barrier for electron transport is roughly 1–1.5 eV (as determined from a Fowler analysis¹⁵), the photon energy is, in principle, high enough to create a hole photocurrent for $h\nu > 2.7$ eV. However, when switching between front and back illumination, the amount of absorbed photons will decrease in the top electrode, and increase in the bottom one. Since in Fig. 4, for excitation from the electrodes ($h\nu < 4.2$ eV), the positive contribution decreases and the negative contribution increases, this directly shows that, for this low-energy regime, a negative photocurrent originates from *electrons* excited in the bottom electrode and not from *holes* excited in the top electrode. Similar reasoning holds for a positive photocurrent. Apparently holes play no significant part in this low energy photocurrent contribution.

The low-energy photocurrent is therefore a summation of only electrons excited in both the top and bottom electrodes, which traverse the barrier in opposite directions. The sign of the resulting net photocurrent depends now on the relative absorption, excitation, transport and transmission of the two electrodes.²³ Choosing the correct experimental conditions, this can give further insight in the quality of the barrier and its interfaces.

In Fig. 5 photoconductance curves for a 1.2 and 1.4 nm Ta layer, oxidized for different oxidation times are presented. The two different energy ranges, below and above the 4.2 eV bandgap, can clearly be discerned. For low photon energies ($h\nu < 4.2$ eV) the net photocurrent is determined by the summation of two opposite electron currents originating from the two electrodes, as discussed above. The resulting sign and magnitude of this part of the photocurrent depend both on transport processes *towards* the barrier, as well as transport *within* the barrier layer. The latter process can possibly be affected by an asymmetric barrier profile, as previously

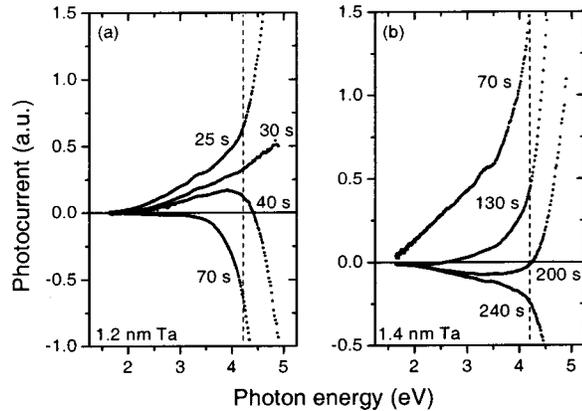


FIG. 5. Photoconductance curves for (a) 1.2 nm Ta and (b) 1.4 nm Ta oxidized for varying oxidation times. The low-energy photocurrent tends toward negative values with increasing oxidation time. A positive photocurrent corresponds to an electron flow from top to bottom electrode. The light is incident on the top electrode and no bias voltage is applied.

shown for underoxidized AlO_x MTJs.⁵ For high ($h\nu > 4.2$ eV) photon energies the sign of the additional photocurrent is completely determined by the barrier asymmetry,²² which is given in Fig. 1(c).

For both Figs. 5(a) and 5(b) the low-energy as well as the high-energy photocurrent is seen to decrease and become negative with increasing oxidation time. This suggests that the sign-change of the barrier asymmetry, as shown in Fig. 1(c), also influences the low-energy photocurrent. However, if we compare the sign of the low-energy photocurrent for the “optimal” oxidation times (i.e., $\Delta\phi=0$) of Figs. 5(a) and 5(b) a clear distinction can be seen. For a 1.2 nm (and thinner) Ta barrier layer the low-energy photocurrent is in this case (30 s oxidation) positive, while for a 1.4 nm Ta layer (220 s oxidation) it is already negative. The overall efficiency of the processes in the top electrode that lead to hot-electron transport through the barrier (photoexcitation, hot-electron transport to the barrier, and interface transmission) thus decreases with respect to that of analogous processes in the bottom electrode. This relative hot-electron efficiency decrease can be attributed to the structural changes near the barrier interfaces with increasing Ta thickness, for example caused by s - d interfacial scattering.¹³ Similarly, these structural changes may cause the observed decrease of the MR ratio with increasing Ta thickness.

V. CONCLUSIONS

In this paper TaO_x has been investigated as a possible alternative barrier material in MTJs. The significantly lower barrier height, as compared to the mostly used AlO_x , would make it an attractive option for low $R \times A$ junctions. How-

ever, the magnitude of the MR ratio is significantly lower and shows a strong decrease with increasing Ta layer thickness. Current-voltage measurements and structural characterizations show evidence for nonhomogeneous oxygen growth. From photoconductance experiments accompanying changes in the hot-electron transport into the barrier have been found, indicative of a structural modification. Such extrinsic processes can adversely affect the spin-polarized tunnel transport. However, the influence of intrinsic bandstructure effects cannot be excluded.

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