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Oxidation process of AlO_x-based magnetic tunnel junctions studied by photoconductance

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The oxidation process of $Co/AlO_x/Co$ magnetic tunnel junctions has been investigated by photoconductance, in addition to traditional transport measurements. The shape of the photoconductance curves is explained within the framework of a simple qualitative model, assuming an oxidation time dependent imbalance of the incident forward and reverse hot electron fluxes, as well as inelastic scattering processes in the oxide. Due to the large sensitivity of the technique, the presence of unoxidized Al beneath the barrier layer can be monitored very accurately. The disappearance of a negative contribution to the photocurrent indicates the complete oxidation of the barrier layer, which coincides with the maximum magnetoresistance. From a Fowler analysis, the barrier height is determined as a function of oxidation time. The observed disagreement of the effective barrier heights determined by this technique and those found by Simmons fits demonstrates the added value of photoconductance studies. © *2003 American Institute of Physics.*
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In the field of spin-electronics, magnetic tunnel junctions (MTJs) are of great interest for potential applications such as sensor elements in hard-disk heads and as nonvolatile memory elements.[1] A crucial step in the fabrication of MTJs is the formation of a thin insulating barrier layer. Despite the great amount of work in searching for alternative barriers, aluminum oxide is still the most promising and widely used barrier material. The AlO_x barrier is generally formed by plasma oxidation of a thin Al layer. Junctions containing suboptimally oxidized barriers can show a severely reduced tunnel magnetoresistance (TMR), due to a loss of spin polarization of the tunneling electrons. Several techniques have previously been used to investigate the oxidation process.[2–4] However, most of these techniques use special samples or incomplete junctions, and could therefore not be *directly* related to other junction properties. Previously we have shown that Al can be used as an efficient source for photoexcited hot electrons.[5] Therefore photoconductance is a first-choice technique to study the gradual oxidation of a thin Al layer in detail. In this paper we use this technique to investigate the oxidation process of the Al-layer ''afterwards'' in complete MTJs. A comparison is made with results from traditional transport measurements.

The junction structures studied were prepared by UHV dc magnetron sputtering through metal contact masks onto glass substrates, resulting in $200\times200\ \mu m^2$ junction areas. The details of this preparation technique have been given elsewhere.[5] All MTJs studied in this work have the same generic structure: glass//3.5 Ta/3.0 $Ni_{80}Fe_{20}$/10.0 $Ir_{20}Mn_{80}$/2.5 $Ni_{80}Fe_{20}$/1.5 Co/1.7 Al+oxidation/4.0 Co/10.0 $Ni_{80}Fe_{20}$/3.5 Ta, with all thicknesses in nanometers. The oxidation time of the Al layer was varied from 40 s up to 200 s. Afterwards the junctions were annealed briefly in a magnetic field of 40 kA/m at 300 °C in order to define the biasing direction.

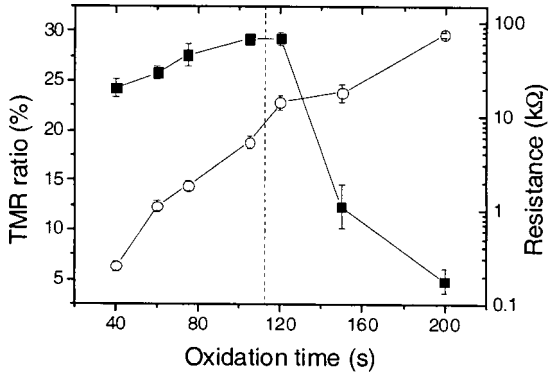

FIG. 1. TMR ratio (■) and parallel resistance (○) for MTJs with different oxidation times for the Al barrier layer. The dashed line corresponds to the oxidation time where the Al layer is completely oxidized.

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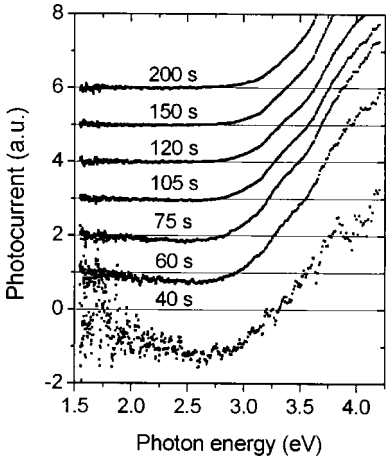

FIG. 2. Photoconductance curves of MTJs with different oxidation times. All curves are measured with zero applied voltage and with the light incident on the top electrode. The curves have been shifted for clarity. The thin lines correspond to the zero current value.

For the conductance, magnetoconductance, and photoconductance measurements, all carried out at room temperature, a homebuilt setup has been used.[5]

In Fig. 1 the TMR ratio and the junction resistance for different oxidation times are presented. The junction resistance shows (roughly) an exponential increase with oxidation time, which is related to the growth of the barrier layer.[2] The TMR ratio, however, shows a maximum around an oxidation time of 105–120 s. Such a maximum in the TMR has also been observed by others,[2,6,7] and has been attributed to the full oxidation of the Al layer, without oxidizing the underlying magnetic electrode.

The *same* junctions were also characterized by photoconductance to study the oxidation process in more detail, as shown in Fig. 2. The curves were measured with zero voltage applied over the junction and with light incident on the top electrode. The shape of these curves can be understood qualitatively from the observed photoconductance curves for two model junctions, shown in Fig. 3, for either light incident on

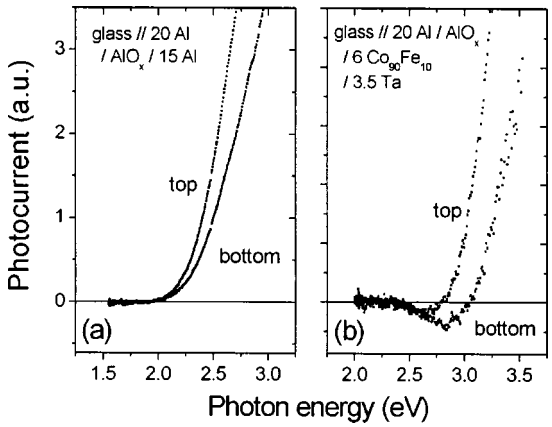

FIG. 3. Photoconductance curves for top and bottom illumination in two model junction structures, showing (a) the domination of the photocurrent by electrons excited in the top electrode, and (b) the presence of a negative photocurrent contribution, due to the large photoexcitation efficiency of Al. The thicknesses are in nanometers.

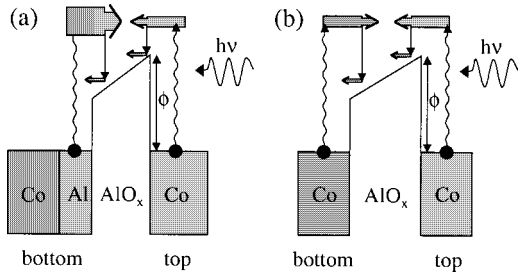

FIG. 4. Energy diagrams showing the photoexcitation of hot electrons and their scattering in the AlO_x barrier layer, for (a) underoxidized and (b) optimally oxidized MTJs. The light is incident through the top electrode. The size of the gray arrows indicates the relative amplitude of the hot electron currents.

the top or the bottom electrode. A positive photocurrent corresponds to a net flow of electrons from top to bottom, and a negative photocurrent to the reverse situation. For a junction with two Al electrodes [Fig. 3(a)], a positive photocurrent is observed, regardless of the direction of the incident light beam. From a simple optical calculation (not shown) it is deduced that for bottom illumination the amount of excited electrons near the interface is larger in the bottom compared to the top electrode. Since no negative photocurrent is observed for bottom illumination, this implies that electrons excited in the top electrode have a higher chance of reaching the opposite electrode, compared to electrons excited in the bottom electrode. This difference is conjectured to be due to inelastic scattering in the built-in field in the oxide conduction band.[8] The current due to electrons excited in the bottom electrode and traversing the oxide layer opposite to the built-in field will be attenuated. In contrast, electrons excited in the top electrode will reach the opposite electrode in spite of such processes. This means that the net photocurrent will be dominated by electrons excited in the top electrode. For the junction structure with an Al bottom electrode and a magnetic top electrode [Fig. 3(b)], a negative photocurrent contribution is observed for small photon energies. Since this negative contribution is not present for a structure with identical electrodes, it is concluded that Al is more efficient than $Co_{90}Fe_{10}$ in generating hot electrons that contribute to the photocurrent. While switching from top to bottom illumination the curve does not only change in amplitude [like Fig. 3(a)], but also the shape itself changes. This demonstrates clearly the two separate contributions to the net photocurrent.

The photoconductance measurements for the MTJs for different oxidation times (Fig. 2) can now be interpreted as illustrated in Fig. 4. If we assume that the mean-free path for hot electrons is quite small compared to the optical penetration depth and the thickness of the Co layers,[9] and that AlO_x is optically transparent in the photon-energy range used, equal amounts of electrons will be excited in the top and bottom electrode. However, due to the oxidation process an asymmetrical potential barrier shape will form,[10] resulting in an internal field, which accelerates electrons in the conduction band of the oxide in the downward direction (see above). This means that, under the above assumptions, the net photocurrent will be dominated by electrons excited in the top electrode. This explains the results for optimal and

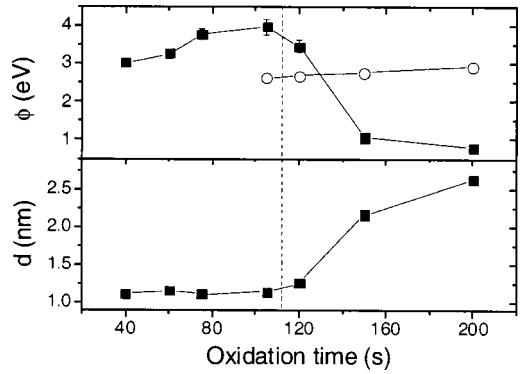

FIG. 5. Barrier height ϕ and barrier thickness d for different oxidation times, as determined from a Simmons fit to the current–voltage characteristics (■). In the upper graph ϕ as determined from the photoconductance curves (○) for optimal and overoxidized junctions is also shown.

overoxidized junctions. However, as illustrated in Fig. 3(b), Al is more efficient in generating photoelectrons than $Co_{90}Fe_{10}$. Within a three-step model for the internal photoemission process (excitation-propagation-transmission) this can be viewed as being caused either by a difference in the optical excitation rate to final states from which the transmission probability through the barrier is high, and/or by a difference in the hot electron scatter rate during propagation to the barrier. This means that for oxidation times where the Al is not completely oxidized a highly efficient source of hot electrons is present just under the barrier, which generates a strong negative contribution to the photocurrent. Due to an increase of the scatter rate in the AlO_x conduction band with energy,[11] the negative contribution decreases at higher energies, while the (positive) photocurrent contribution from the top electrode will continue to increase. This will cause a sign reversal of the photocurrent for higher energies. The disappearance of the negative photocurrent contribution between 105 and 120 s of oxidation is therefore interpreted as the time at which the Al layer is completely oxidized. Indeed the TMR ratio is then found to be optimal.

From a fit using the Simmons model[12] to the current–voltage characteristics, the barrier height ϕ and thickness d have been deduced. In Fig. 5 we compare these values with ϕ as determined from a Fowler analysis[8] of the photoconductance curves for optimal and overoxidized junctions. For the underoxidized junctions a more complex model would be needed to unravel the contributions from the two electron fluxes, and to be able to extract an effective barrier height. First, the bottom to top electron current contribution is not simply proportional to $(h\nu-\phi)^2$, due to the energy dependence of scattering processes (see above). Second, for the underoxidized junctions the (negative) photocurrent starts already at a lower photon energy compared to the optimal and overoxidized ones. This is either an indication of the effect of photon-assisted tunneling through the top of the potential barrier,[13] a lower barrier height due to the incomplete formation of the barrier, or a combination of these two.

The barrier height ϕ as determined from the Simmons fit shows a maximum around 105 s, followed by a rapid decrease. Qualitatively a similar behavior was also measured by van de Veerdonk.[14] However, the barrier height as determined from the photoconductance curves shows a weak increase towards higher oxidation times. This indicates that either the Simmons model is not appropriate for these MTJs, or that the effective barrier height for the tunneling electrons differs from that for photoexcited electrons. It should be noted that in the Simmons model the barrier is assumed to be rectangular,[12] whereas the value of ϕ as deduced from a Fowler analysis is assumed to coincide with the maximum of the barrier. For overoxidized junctions it has been suggested that Co is being incorporated within (as impurities) or underneath (as CoO_x) the barrier.[14] The resulting composite barrier can then support thermally and field-assisted electron transport via impurity states. A Simmons fit then leads to an effective barrier height which is lower than the maximum distance from the Fermi level to the bottom of the conduction band. However, conclusive evidence of this supposition for our MTJs will require further investigation.

Summarizing, in this paper we have demonstrated the viability of the photoconductance technique as a characterization tool for the oxidation process of *complete* MTJs. The changes in the photoconductance curves with oxidation time are explained within the framework of a simple qualitative model. Due to the large sensitivity of the technique to the presence of Al close to the tunnel barrier, the disappearance of a negative contribution to the photocurrent can be correlated to the complete oxidation of the barrier layer and the maximum in TMR. An observed difference between the barrier height as determined by Simmons fits and by photoconductance is indicative of the added value of photoconductance over traditional transport measurements. The high potential of this technique will be used in forthcoming work to study the dependence on applied voltage, the influence of anneal conditions, as well as the use of alternative barrier materials.

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