Ultraviolet light assisted oxidation for magnetic tunnel junctions

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Recently, ferromagnetic tunnel junctions attracted much interest because they exhibit large magnetoresistance effects up to $\Delta R/R = 40\%$ at room temperature. In this paper, an alternative approach will be presented, i.e., exposing the aluminum film to ultraviolet light during oxidation. The underlying mechanism can be explained in the framework of the Cabrera–Mott theory of oxidation. With UV assisted oxidation, magnetoresistance ratios of up to 22.5% at room temperature have been obtained in $Ni_{80}Fe_{20}$–Al(ox)–Co junctions. The resistivities were in the 1 k$\Omega\,\mu$m^2 range for junctions of 25–400 μm^2. Barrier heights obtained by fitting to different theories will be discussed. © *2000 American Institute of Physics.* [S0021-8979(00)55308-0]

I. INTRODUCTION

Over the past years, tunnel magnetoresistance (TMR) attracted much attention as an alternative to the giant magnetoresistance in the design of magnetic field sensors. In addition, magnetic tunnel junctions (MTJ) are possible key devices for future nonvolatile random access memories (magnetic RAM, MRAM). For the latter application, the magnetic tunnel junctions have to compete with state-of-the-art ferroelectric or semiconductor based nonvolatile memories. For highly integrated MRAMs submicrometer sized MTJs are desired. The absolute resistance has to be in the order of kiloohms to match electrically CMOS write and sense circuits. Compared to other magnetoresistive devices the resistance area product ($R \times A$) in tunnel junctions can be adjusted over several orders of magnitude. This is an interesting feature because it offers the possibility to tune the absolute resistance for sensors and nonvolatile memory applications.

Most of today's tunnel barriers are made of aluminum which after deposition is oxidized. Additionally to thermal oxidation in pure oxygen, resulting in very low area resistivities of some 10 $\Omega\,\mu$m^2,[1] oxidation in air[2] or by plasma oxidation (oxygen glow discharge)[3,4] are most often met. The latter two methods give high area resistivities of at least some 100 k$\Omega\,\mu$m^2, unless one applied extremely short and less precise oxidation times. Our method of ultraviolet light assisted oxidation fills the gap by providing area resistivities of 100 $\Omega\,\mu$m^2–10 k$\Omega\,\mu$m^2 in an apparently self-limiting oxidation process.

II. EXPERIMENTAL METHODS

Samples have been grown in a Leybold Z400 sputter equipment with three sputter targets, Co, $Ni_{80}Fe_{20}$, and Al. Both ferromagnets were deposited by dc magnetron sputtering, and aluminum by rf sputtering. The deposition rates are 1.8, 1.6, and 0.3 nm/s, for Co, NiFe, and Al, respectively. Two different ferromagnets have been chosen to ensure that the two different coercive fields lead to an antiparallel alignment of the magnetizations by varying an external magnetic field. MTJs of the form $Ni_{80}Fe_{20}$–Al(ox)–Co and Co–Al(ox)–$Ni_{80}Fe_{20}$ have been prepared and investigated. The bottom electrode served as a contact lead. The $Ni_{80}Fe_{20}$ layers usually were 19 nm thick, the Co layers were 15 nm. The deposited Al thickness varied between 1 and 3 nm.

After deposition of the trilayers, tunnel junctions were patterned using a self-aligned process which requires three mask steps.[4] Structures were defined by optical lithography and etching was done with argon ion beam etching. In the first step, the complete trilayer was shaped into the form of the bottom electrode. Second, the junctions were defined by argon ion milling, which was stopped on reaching the bottom layer. The etch process was *in situ* analyzed by secondary ion mass spectrometry (SIMS). Immediately after etching, a SiO passivation layer has been evaporated. After lift-off of the SiO, the top gold wiring layer was patterned also with the lift-off technique.

Before processing, the samples have been characterized by magneto-optical Kerr effect (MOKE) measurements. Current–voltage and magnetoresistance measurements were performed in a computer controlled setup at temperatures from 10 K to room temperature after the junctions had been completely processed.

III. OXIDATION

The influence of ultraviolet light on oxidation first has been studied by Cabrera[5] in the framework of the Cabrera–Mott theory of oxidation.[6] Experimentally, ultraviolet light has only sparsely been used to support oxidation, e.g., by Boyd *et al.* for silicon,[7] and by Fritzsch and co-workers[8] for niobium based Josephson tunnel junctions. Here, it is applied in the oxidation of ferromagnetic tunnel junctions.

According to the Cabrera–Mott theory, during thermal oxidation in pure oxygen, electrons tunnel from the metal through the already formed oxide layer to adsorbed oxygen atoms. The reason for this is that the lowest unoccupied oxygen level lies deeper than the Fermi energy of aluminum. This builds up an electric field across the oxide layer. Metal

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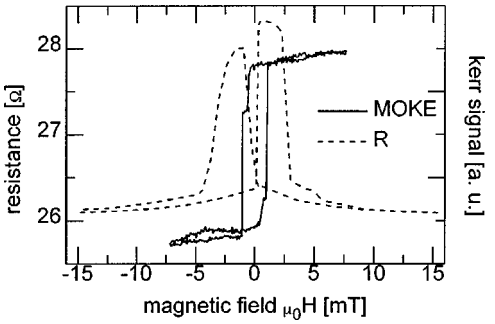

FIG. 1. MOKE hysteresis loop and magnetoresistance effect at 300 K of a sample with 1.7 nm barrier thickness and junction size of 25 μm^2. The magnetoresistance effect here is about 8.5%.

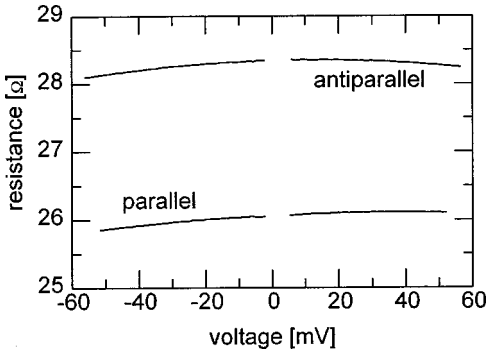

FIG. 2. Resistance vs voltage for parallel and antiparallel magnetization alignment for the same sample as in Fig. 1.

ions at the metal–oxide interface are pulled by this field through the oxide to the surface. These metal ions form together with the oxygen the oxide, i.e., the tunnel barrier. If the field becomes weaker due to the increasing oxide thickness, the oxide growth slows down and eventually stops. The ultraviolet light enables more electrons to cross the oxide through the photoelectric effect. This builds up a stronger electric field so that the final oxide layer is thicker.

For our experiments, a 4 W low pressure mercury lamp was mounted into the chamber. During oxidation, the lamp is positioned to shine light onto the samples. Thus, the complete trilayer is fabricated without breaking the vacuum. In addition to ultraviolet light supported oxidation, some samples have been oxidized *ex situ* in air or *in situ* in pure oxygen. For *in situ* oxidation, a pressure of 100 mbar has been used, for 15 min to 3 h.

IV. RESULTS

The MOKE characterization was used to determine the magnetic properties of the trilayers. All samples with an oxidized tunnel barrier showed a separate switching of both ferromagnets. Yet, the switching fields were closer together than would be expected without coupling from hysteresis loops of single metal layers of the same thickness. This indicates an orange peel coupling. If the ferromagnetic layers are separated by a 1.3 nm unoxidized aluminum layer, they switch at once, whereas in the case of 3 nm interlayer thickness a separate switching was observed.

Figure 1 shows both a MOKE hysteresis loop and a TMR measurement of the same trilayer. While the soft magnetic bottom $Ni_{80}Fe_{20}$ layer has the same switching field in both measurements, the Co switching field increased in the TMR measurement. For junctions with a larger area, this effect was less pronounced. We suppose that the smaller layer size of the top electrode reduces the domain wall mobility.

For samples of nominally 1.0–1.3 nm barrier thickness, the area resistivities range between 200 and 1500 $\Omega\,\mu$m^2. The magnetoresistance effect usually is about 10%–13% at room temperature; a maximum of 22.5% has been found. If the aluminum layer becomes thicker than 1.3 nm, the magnetoresistance effect starts to decrease. The exposure time during oxidation between 15 min and 3 h had no noticeable effect on the junction properties.

Some samples were prepared by oxidation in ambient air or in pure oxygen. The latter had high area resistivities of about 1 M$\Omega\,\mu$m^2. Due to the thick Al layer, 3 nm and more, the magnetoresistance ratio was below 3%. The samples oxidized in pure oxygen had very low area resistivities, such that geometry effects made an accurate determination of area resistivity and magnetoresistance ratio impossible. The area resistivities can be estimated to lie in the order of 10 $\Omega\,\mu$m^2.

The current–voltage characteristics exhibit the typical non-Ohmic tunnel behavior; with increasing voltage the tunnel resistance decreases (Fig. 2), and also the magnetoresistance effect (Fig. 3). These results are in qualitative agreement with typical measurements on plasma oxidized samples. In Fig. 2, the Brinkman formula[9] has been fitted to the current–voltage curve for parallel alignment. Due to the asymmetry of the $I(V)$ curve, Simmons' formula[10] seems not to be appropriate for fitting because he replaced the barrier shape by a mean barrier height thus eliminating the effect of a barrier asymmetry. The values for the mean barrier height and the barrier asymmetry are $\varphi=1.03$ eV and $\Delta\varphi=0.53$ eV, the barrier thickness is $s=12.3$ Å. As one should expect, the sign of $\Delta\varphi$ changes if the ferromagnets are exchanged; the resistivity is higher if electrons go from $Ni_{80}Fe_{20}$ to Co. This means that the work functions of permalloy electrons into the barrier conduction band is higher than those for cobalt electrons. From the work function of Ni (5.15 eV), Fe (4.5 eV), and Co (5.0 eV) against vacuum, one might assume in spite of all problems that come with work functions, that the $Ni_{80}Fe_{20}$ work function also is higher than that of Co which c would support the above measurements. A closer agree-

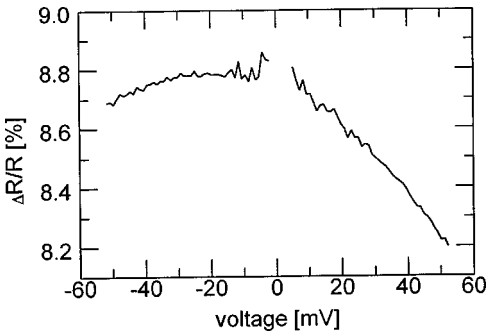

FIG. 3. $\Delta R/R$ extracted from Fig. 2.

ment between theory and experiment cannot be expected because in the development of these and most other fit formulas, some assumptions were made[11] which eliminate any dependence on the band structure of the metal electrodes. In contrast, the existence of magnetoresistance is a consequence of the ferromagnetic band structure.

V. SUMMARY

It has been shown that ultraviolet light is able to support the oxidation of aluminum in the formation of tunnel barriers for ferromagnetic tunnel junctions. The resulting oxide layer is free of pinholes which has been verified by the existence of a tunnel characteristic and a tunnel magnetoresistance of 10% and above. The area resistivities on the order of 1 $k\Omega\,\mu m^2$ are higher than those obtained by thermal oxidation in pure oxygen, but more than an order of magnitude lower than the results of the other two common oxidation methods, oxidation in air and plasma oxidation. Thus, the use of ultraviolet light during oxidation fills a gap in area resistivities which existed between the most common oxidation procedures.

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