Dielectric breakdown of ferromagnetic tunnel junctions

W. Oepts, a) H. J. Verhagen, and W. J. M. de Jonge
Department of Physics and COBRA, Eindhoven University of Technology, 5600 MB Eindhoven, The Netherlands

R. Coehoorn
Philips Research Laboratories, Prof. Holstlaan 4, 5656 AA Eindhoven, The Netherlands

(Received 26 May 1998; accepted for publication 18 August 1998)

The time-dependent dielectric breakdown of Co/Al$_2$O$_3$/Co(-Fe) magnetic tunnel junctions is investigated. At voltages larger than 1.2 V, almost immediate breakdown of the junction is observed, leading to a decreased (magneto)resistance. The shorts, which are local hot spots, were visualized by making use of a liquid crystal film on top of the junction. The breakdown voltages of a series of nominally identical tunnel junctions measured in a voltage-ramp experiment are shown to increase with increasing ramp speed. The results are analyzed in the framework of several models for the voltage dependent breakdown probability. © 1998 American Institute of Physics.

Recently, a large magnetoresistance (MR) effect has been found for tunnel junctions consisting of two ferromagnetic electrodes separated by a thin oxidic barrier. 1 This so-called tunnel MR effect occurs upon a change of the relative orientation of the magnetizations of the two electrodes in an applied field, and is the result of spin-dependent electron tunneling. These junctions are potentially applicable in magnetoresistive heads, magnetic field sensors, and magnetic tunneling. Crucial for the successful operation of a magnetic tunnel junction are the properties of the insulating barrier, which must be pinhole free with a thickness $d$ of 1.0–2.0 nm in order to have a sufficiently high tunneling probability. Due to the small barrier thickness, applied voltages of only 1 V lead to electric fields of the order of $E \approx 1 \times 10^9$ V/m. Above these electric fields dielectric breakdown of oxides is commonly observed. 2,3 We have observed indeed that breakdown of representative Co/Al$_2$O$_3$/Co tunnel junctions, with $d \approx 2.0$ nm, occurs almost immediately upon the application of voltages above 1.2 V. In order to assess the applicability of these junctions for devices which operate at much lower voltages, we have investigated their time-dependent dielectric breakdown by measuring their lifetime in a series of voltage ramp experiments. As a first step towards the understanding of the microscopic mechanism of breakdown we have analyzed the results in terms of various expressions for the voltage-dependent probability of breakdown.

We fabricate our junctions by using an in situ shadow mask system, with a cross-bar geometry. In one run we are able to grow 64 junctions with varying electrode widths of 50, 100, 150, and 200 $\mu$m. First a bottom electrode consisting of 20 nm of Co is evaporated through the shadow mask on a liquid N$_2$ cooled insulating Si(100) substrate. Evaporation is carried out in an UHV Balzers UMS630 multichamber molecular beam epitaxy (MBE) system, at a base pressure in the $10^{-8}$ Pa ($10^{-10}$ Torr) range. Thereafter the substrate is transported to a separate UHV chamber ($P_{\text{base}} = 1 \times 10^{-7}$ Pa), and without a shadow mask the thin Al layer is sputtered from a 2 in. Al target, purity 99.999%, in an Ar pressure of 0.6 Pa. Immediately after deposition the chamber is pumped down and then filled with O$_2$ (99.999% purity) to a pressure of 9.2 Pa. A dc glow discharge is ignited from a ring-shaped cathode at a voltage of $-1.6$ kV with respect to both substrate and UHV chamber. In 100–300 s the Al layer is oxidized. After pumping down, a 20–80 nm Co or Co$_{50}$Fe$_{50}$ top electrode is evaporated in the MBE system, again through a shadow mask.

Our fabrication process leads to junctions with resistances varying from 1 to 500 k$\Omega$ (for the lateral dimensions mentioned), depending on the Al layer thickness and oxidation time. The resistance of junctions with the same area was observed to be equal within 10%. In Fig. 1 a typical $I$–$V$ characteristic of a junction is shown. Initially the current increases linearly with the voltage, and at larger voltages an exponential increase is seen which is in agreement with Simmons’ theory of tunneling. 4 Using Simmons’ model we obtained, for this junction, values for the barrier height $\phi = 1.0$ eV and barrier thickness width $d = 2.2$ nm. In the inset a MR curve of a Co/Al$_2$O$_3$/Co$_{50}$Fe$_{50}$ junction is shown, with a MR ratio of 18%. The absence of a clear high resistance

FIG. 1. Current–voltage characteristic of a Co/Al$_2$O$_3$/Co$_{50}$Fe$_{50}$ junction. The dashed curve is an enlargement of $100 \times$ the measured curve, at 1.17 V a breakdown occurred. The inset shows the MR curve for a 150×50 $\mu$m$^2$ junction at room temperature.
plateau indicates that a perfect antiparallel state of both electrodes is not quite reached in this case. The tunnel junctions showed a MR ratio varying from 5% to 18%, which decreased with increasing $V_{bias}$, typically 50% at $V_{bias} = 0.3$ V. The junctions used for the series of voltage breakdown measurements discussed below are all grown in the same run, with a Co(20 nm) bottom electrode of various widths and a 50-$\mu$m-wide Co (80 nm) top electrode. The fitted barrier parameters are: $d = 2.0$ nm and $\phi = 1.0$ eV. Some junctions had a very low resistance and showed no MR, or had a lower resistance but showed breakdown on a much shorter time scale than its neighbor tunnel junctions. These atypical junctions, as well as the junctions that broke down during handling, have not been used for the study discussed below.

We observed that breakdown of our junctions occurs typically within minutes when they are operated at a $V_{bias}$ of 1 V. Biasing the junction at 800 mV immediately increased the lifetime to several days. Junctions grown in the same run with the same dimensions showed a comparable lifetime when biased at the same $V_{bias}$. Upon breakdown an almost ohmic $I-V$ curve and no MR effect is observed. After breakdown, resistances were typically of the order of 100–10 $\Omega$. The lowering of the resistance can be understood as the formation of a microscopic ohmic short in the barrier at the moment of breakdown.

With the use of nematic liquid crystal (LC), we were able to visualize the location of breakdown. A thin film LC (ROCE 1540 from Hoffmann LaRoche) dissolved in acetone is deposited on the sample. Upon heating the LC to above the clearing point ($T_c = 56.5$ °C), the LC material changes from the nematic state showing optical anisotropy to the isotropic state. Using linearly polarized light, the state of the LC from the nematic state showing optical anisotropy to the isotropic state. Using linearly polarized light, the state of the LC can be determined: a rise in temperature to above $T_c$ results in the LC film becoming opaque. By operating the junction just below $T_c$, hot spots on the junction surface are then visualized as black spots. In Fig. 2 a CoAl$_2$O$_3$/Co$_{50}$Fe$_{50}$ junction after breakdown is shown, having a hot spot in the center area of the junction surface when operated. The size of the observed spot does not relate to the diameter of the short created. Most junctions showed only one hot spot after breakdown and additional spots could be created by raising the voltage far above 1 V. Approximately half of the junctions investigated had a breakdown close to the edge of the bottom electrode, the remaining junctions showed breakdown randomly distributed over the surface.

For a series of junctions grown in the same run, we have measured the time to breakdown upon ramping the applied voltage from $V = 0$ with a constant $dV/dt$. In Fig. 3 it is seen that $V_{bd}$ increases when the ramp speed ($dV/dt$) is increased, which is in agreement with the observation that at lower $V_{bias}$ the lifetime increases.

We can compare breakdown of our tunnel barriers with the breakdown of Al$_2$O$_3$ or SiO$_2$ based capacitors. As early as 1968 breakdown fields of plasma oxidized Al films for capacitors with varying thicknesses were reported. An $E_{bd}$ of $1 \times 10^6$ V/m for a 1.5 nm film was found, which is comparable to our values of $V_{bd} = 1$ V in a 1.0–2.0 nm barrier. In the past three decades, the mechanism of breakdown has been intensively investigated for MOS capacitors. Investigations of SiO$_2$ breakdown have mostly been carried out on 5–15 nm dielectric films with applied voltages usually larger than the barrier height (in eV) of the insulator, where tunneling is in the Fowler–Nordheim regime. In our studies the applied voltage is relatively small, and only for the highest voltages applied ($\geq 1$ eV) are the electrons expected to enter the conduction band of the insulator. For breakdown of SiO$_2$ dielectrics several models have been proposed, of which the so-called “E model” and the “1/E model” ($E = V/d$ is the applied electric field) are most often applicable. These models differ in both the physical mechanism and the voltage dependence of breakdown. As we have limited data and are uncertain about the mechanism of breakdown in our junctions, we will compare our data with both models.

The $E$ model follows from the assumption that breakdown is related to the field induced displacement of ions in the barrier. By the presence of the field, the effective activation $E_A$ energy for a jump of an ion over a distance $a$ in the direction of the applied field is decreased to $E_A - eaZe/|a|$, where $eZ$ is the ionic charge. The resulting rate of breakdown at an applied voltage $V$ is then given by

$$P = A \exp \left( \frac{V}{E} \right) \quad \text{(for } V \gg B),$$  

where $P$ is the breakdown rate, $A$ is a constant, $V$ is the voltage, and $E$ is the effective activation energy. The result is consistent with the model of breakdown dependence on the voltage and the field.

FIG. 2. Polarized light picture of liquid crystal on top of a CoAl$_2$O$_3$/Co$_{50}$Fe$_{50}$ junction, the black spot in the middle of the junction surface is the location of a breakdown.

FIG. 3. Measured $V_{bd}$ as a function of ramp speed. All measured junctions are grown in the same run. The straight line is the calculated $V_{max}$ for the 50 $\mu$m bottom electrode junctions, the dotted lines indicate the width of the breakdown rate peak vs $V_{max}$.
where the prefactor $A$ is independent of $V$, and proportional to the junction area and to $\exp(-E_A/kT)$ and in which $B = (kT/eZ)(d/a)$. We have studied breakdown in a voltage ramp experiment with constant $dV/dt$. It can be derived from Eq. (1) that the maximum breakdown rate for a large ensemble of identical junctions varies linearly with $\ln(dV/dt)$:

$$V_{max} = B \ln \left( \frac{1}{AB} \right) + B \ln \left( \frac{dV}{dt} \right).$$  (2)

The measured results for $V_{bd}$ for the 50 $\mu$m bottom electrode junctions as given in Fig. 3 were fitted using this equation (full line). We find values of $A = 6.3 \times 10^{-17}$ s$^{-1}$ and $B = 0.035$ V. Additionally, an estimation for the statistical variation is given by the dashed lines, which connect the inflection points of the rate distribution function. The measured $V_{bd}$ values all fall within the interval in between the dashed lines, which is consistent with the model used. We can crudely estimate the parameter $B$ by assuming that the ion displacement takes place by $\text{Al}^{3+}$ ions ($Z = 3$), $kT = 0.025$ eV (room temperature), $a = 0.2$ nm (distance between atomic sites), and $d = 2$ nm, which gives $B = 0.083$ V. This value is of the same order as the observed value.

The $1/E$ model follows from the assumption that during electrical stress trap states are created in the barrier, giving rise to additional conduction paths for electrons. For the case of MOS capacitors with a SiO$_2$ dielectric layer it has been assumed that traps are created by a hole current into the dielectric layer. The holes are formed in the positive electrode as a result of electron excitation from a state below the Fermi level by the highly energetic electrons that arrive after tunneling. The breakdown rate has an $\exp(-1/V)$ dependence, which follows from taking into account that upon increasing the voltage, the probability of a hot electron generating a hole increases. The probability of breakdown according to the $1/E$ model is

$$Pdt = C \exp \left( -\frac{D}{V} \right) dt,$$  (3)

in which the prefactor $C$ is proportional to the junction area and to the current density, because more holes will be generated at a higher current density. $D$ is a constant depending on the material properties and barrier thickness. Again we can express $V_{max}$ in terms of $dV/dt$ and use this to fit the data in Fig. 3. The assumption that $C$ is the product of a constant and the experimentally determined, voltage dependent, current density did not lead to a consistent fit. However, assuming that $C$ is just a constant gave a good fit to the data, with $C = 5.6 \times 10^9$ s$^{-1}$ and $D = 31$ V. Of course, the necessity to assume that $C$ is independent of $V$ does not provide strong support for the applicability of the $1/E$ model.

The extrapolated lifetimes at a constant lower voltage, as obtained within both models, differ largely. At 0.5 V we find within the $E$ model and the $1/E$ model a lifetime of 217 and 10$^8$ yr, respectively. The use of both models suggest that the lifetime is sufficient for practical applications at voltages $\leq 0.5$ V. However, referring to observed deviations from these models at low voltages for SiO$_2$ capacitors, we emphasize that more extensive low voltage studies are still essential. From Fig. 3 it is seen that $V_{bd}$ does not significantly decrease with increasing junction area, which would be expected within both models. The resistance of these junctions decreased by only 50% when the bottom electrode width increased from 50 to 200 $\mu$m, suggesting a nonuniform current density distribution across the junction area. This cannot be the result of a geometrical artefact, since the junction resistance ($\sim 100$ $\Omega$) is much larger than the sheet resistances of the electrodes ($\sim 10 \Omega$). A lower resistivity at the edges can be a possible explanation. With the use of an atomic force microscope it was found that, due to the deposition through a shadow mask, the bottom electrode edge is approximately 10 $\mu$m broad. Assuming a differing resistivity for the edge surface and remaining junction surface, the resistance scaling can then be explained with a six times lower edge resistivity compared with the remaining surface resistivity. The observed enhanced probability of breakdown at these edges supports this assumption.

In summary, we observed almost immediate breakdown in successfully fabricated magnetic tunnel junctions with a plasma oxidized Al$_2$O$_3$ barrier of 2.0 nm when a voltage larger than 1.2 V is applied. It is known that AMR and GMR read heads can also be damaged by electrostatic discharge, but typical voltages leading to damage are much higher, viz. tens of volts. In order to come to an applicable device, with a low tunnel resistance, a barrier thinner than 2.0 nm is needed, and thus the value of $V_{bd} \approx 1$ V seems to be inherent to this device. At the moment of breakdown a microscopic short is created, of which the exact nature and mechanism for creation is still uncertain. The results of a series of field-dependent breakdown measurements can be described well within the so-called $E$ model. Extrapolation of the lifetime to realistic low operation voltages ($V < 0.5$ V) suggest that, if accidental peak voltages outside this region can be avoided, breakdown will not be a limiting factor upon applying these junctions in sensor or MRAM devices.

The authors thank M. Zellenrath for help with the junction fabrication and D. B. de Mooy and V. Zieren for the LC experiments. This work was supported by the European Community ESPRIT Research Project ‘‘Novel Magnetic Nanodevices of Artificially Layered Materials’’ (NM2)².

---

9. Our approach leads to a similar equation as the approach leading to Eq. (8) of B. Schindl, J. S. Suehle, C. Messick, and P. Chaparala, IEEE Int. Reliability Phys. Proc. 34, 84 (1996).

---

**Notes:**