Suppression of orange-peel coupling in magnetic tunnel junctions by preoxidation

W. F. Egelhoff, Jr.,[a] R. D. McMichael, C. L. Dennis, M. D. Stiles, A. J. Shapiro, B. B. Maranville, and C. J. Powell
National Institute of Standards and Technology, Gaithersburg, Maryland 20899

(Received 6 December 2005; accepted 10 March 2006; published online 21 April 2006)

We have found that preoxidation of the bottom Co electrode in magnetic tunnel junctions (MTJs) very effectively suppresses orange-peel coupling. The result is a free layer that is much softer. Work by others has demonstrated that preoxidation is compatible with high-quality MTJ fabrication. [DOI: 10.1063/1.2198087]

A few years ago, it was reported that preoxidation of the bottom Co electrode in magnetic tunnel junctions could dramatically suppress the intermixing that occurs when Al is deposited on Co.[1] It was also found that this preoxidation procedure not only increased the tunneling magnetoresistance (TMR) but dramatically reduced the scatter of data points in a plot of TMR vs *RA* product, where *R* is the resistance of the magnetic tunnel junction (MTJ) and *A* is the tunneling area.[2] The observation that preoxidation suppresses intermixing has been confirmed and extended in recent work.[3] Also reported a few years ago was the observation that in giant magnetoresistance (GMR) spin valves, the oxidation of the surface of the top Co layer suppressed magnetic poles at bumps in the surface.[4] The data suggested that bumps oxidize first and the system self-passivates as a flat oxide-metal interface forms. In light of these observations, it seemed likely that preoxidation of the bottom Co electrode in MTJs might be an effective way of suppressing orange-peel coupling.

Orange-peel coupling is a common problem when MTJs are used to detect low magnetic fields.[5] As illustrated in Fig. 1, this form of coupling is associated with conformal roughness, which produces magnetic poles that favor parallel alignment and act as a coupling field. This coupling field reduces the sensitivity of the free layer to small magnetic fields.

Figure 2 presents data on the coupling field for three different nonmagnetic spacer layers (SLs) in structures of the type Si(100)/250 nm thermal oxide/20 nm $Ni_{77}Fe_{14}Cu_5Mo_4$/0.5 nm Co/*X* nm SL/0.5 nm Co/2.5 nm $Ni_{77}Fe_{14}Cu_5Mo_4$/0.5 nm Co/10 nm $Ir_{20}Mn_{80}$/10 nm Cu, *X* is the thickness of the nonmagnetic SL. In the free layer, the $Ni_{77}Fe_{14}Cu_5M_4$ is for softness and the Co is for high TMR. The coupling field is measured as the shift from zero field of the center of the easy-axis hysteresis loop. The surface roughness of the thermal oxide substrates is not expected to contribute to the measured coupling fields since it is more than an order of magnitude smaller than that of our films.

In Fig. 2, the three nonmagnetic spacer layers exhibit very different coupling fields. When Cu is used the structure is a spin valve, and the coupling field rises sharply for spacings below 2 nm, as is typical for spin valves. When a TMR structure is made in the conventional manner of depositing metallic Al and subsequently oxidizing it, the coupling is somewhat smaller than in a typical spin valve, but still large enough to be problematic for thicknesses of ≈1 nm that are desirable for MTJs.

A dramatic drop in the coupling field is observed when the Co surface is lightly oxidized before Al_2O_3 deposition. The exposure used for Fig. 1 was 10^{-3} Pa (≈10^{-5} Torr) O_2 for 10 s. However, the exact O_2 exposure is not particularly critical since, while approaching that exposure, the surface is becoming passivated. The Al_2O_3 thickness is estimated using two quartz crystal thickness monitor readings for the Al film thickness and assuming a 29% volume expansion when the Al is oxidized. The uncertainty in this approach is estimated to be ±10%.

The remarkable feature of this preoxidation is that the coupling field scarcely rises as the Al_2O_3 thickness decreases from 5 to 0.5 nm. Clearly, the orange-peel coupling has been suppressed to a remarkable extent. For a thickness below 0.5 nm, the coupling field rises sharply, but this effect is due to pinholes in the Al_2O_3.

The method we use to distinguish between pinholes and orange-peel coupling is based on comparing the coupling field at room temperature and at 77 K. Since orange-peel coupling is magnetostatic and the magnetization of Co is almost the same at the two temperatures, little change in the coupling field is observed. However, the pinning of Co by IrMn increases strongly at 77 K. As a result, pinholes connecting the pinned Co and the free Co result in a strong increase in the observed coupling field upon cooling to 77 K. For more details on this effect, see Ref. 6.

The importance of preoxidation lies in the fact that when the orange-peel coupling is more than a few oersteds, it be-

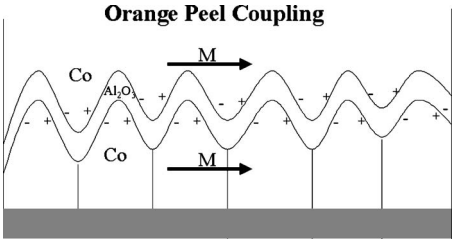

FIG. 1. A schematic illustration of orange-peel coupling based on conformal roughness. The vertical lines represent grain boundaries. The growth is columnar. The plus and minus signs represent the magnetic poles at the bumps on Co. The poles act as a coupling field favoring parallel alignment of the magnetization. The height of the bumps is greatly exaggerated, for clarity, relative to the grain diameters. Note also that in MTJs, grain diameters are typically ten times larger than the Al_2O_3 thickness (Ref. 2).

[a]Electronic mail: egelhoff@nist.gov

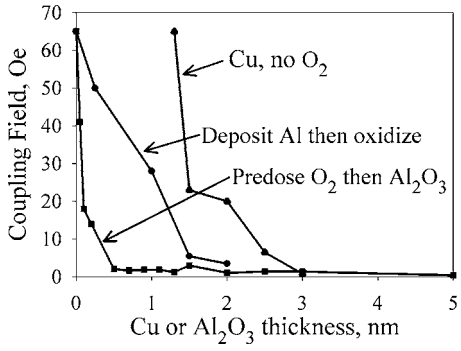

FIG. 2. Plots of the coupling field vs thickness for nonmagnetic (NM) spacer layers in Co/NM/Co structures. The NM spacer layers are Cu with no oxygen used in any way, Al deposited as metal and oxidized subsequently, and preoxidation of the Co before Al_2O_3 deposition.

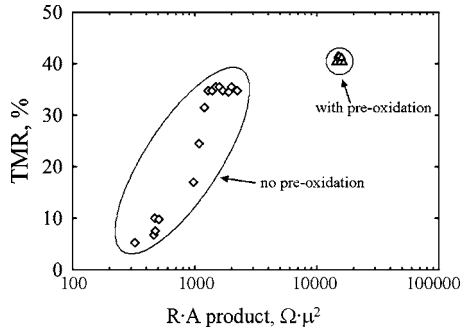

FIG. 4. A plot of TMR vs *RA* product for nominally identical MTJs prepared with and without our preoxidation procedure. The data are a replot of the data in Ref. 2.

comes the dominant contribution to the hard-axis saturation field. This is the axis used in magnetic sensors to achieve linear response. Reducing the orange-peel coupling by an order of magnitude can make a MTJ an order of magnitude more sensitive to small magnetic fields.

Experience suggests that the explanation for the suppression of orange-peel coupling is that given in Fig. 3. It is well established that oxidation of metal surfaces proceeds more rapidly at atomic steps than on flat terraces.[7] The lower coordination of step atoms makes them more reactive and allows oxidation to proceed laterally much more rapidly than vertically down into layers of metal atoms that are fully coordinated. This effect is a manifestation of the effect we reported earlier of bumps oxidizing readily and surfaces self-passivating as a flat insulator-metal interface forms.[4]

As indicated in Fig. 3, such a process has the effect of spreading out the magnetic poles that form at the edges of grains. This spreading out reduces the coupling field sharply, as seen in Fig. 2.

It might be thought that this preoxidation approach would reduce the TMR; however, the available evidence is that it produces an increase in TMR as well as an increase in *RA* product.[2] The data of Ref. 2 are replotted in Fig. 4 for the reader's convenience. The increase in *RA* product may be partly explained by a reduction in interfacial area for tunneling, as Fig. 3(b) would imply. The origin of the increase in TMR is less certain, but may lie in the structure of MTJs at grain boundaries in the absence of preoxidation. It may be that grain boundaries are the site of partial shorts in MTJs, leading to reduced TMR. Note that the Al layer is likely to be thinner near grain boundaries due to diffusion of Al into the grain boundaries.[8] Preoxidation, as illustrated in Fig. 3(b), would eliminate tunneling in regions near grain boundaries.

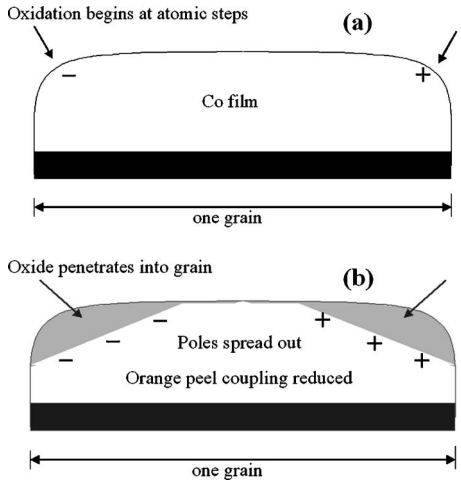

FIG. 3. An illustration (a) of a single Co grain with magnetic poles at the rounded sides and (b) of how oxidation of the rounded sides, because of high atomic-step density, are preferentially oxidized upon exposure to O_2. The effect is to spread out the magnetic poles and to reduce the orange-peel coupling.

The increase in *RA* product with preoxidation is so large that area reductions alone are not likely to be the full explanation. More likely, without preoxidation, partial shorts near grain boundaries reduce the *RA* product.

Another interesting aspect of the model is that it suggests a reason for the marked reduction in scatter of TMR values, as seen in Fig. 4, that is found for nominally identical MTJs.[2] If partial shorts are associated with grain boundaries and preoxidation renders these areas inactive, the tunneling after preoxidation should occur at the relatively flat center regions of each grain that are illustrated in Fig. 3. Such flat regions could easily be expected to give more consistent TMR values than the regions near grain boundaries.

The major conclusions of this work may be summarized as follows. (1) Orange-peel coupling in MTJs may be very effectively suppressed by preoxidation of the surface prior to Al_2O_3 deposition. (2) The suppression of orange-peel coupling allows the sense layer in the MTJ to be softer. (3) The effect is most pronounced for Al_2O_3 thicknesses of 0.5–1.5 nm, which is the range of greatest importance for MTJs. (4) The effect is entirely consistent with large TMR values.

[1]W. F. Egelhoff, Jr., P. J. Chen, R. D. McMichael, C. J. Powell, R. D. Deslattes, F. G. Serpa, and R. D. Gomez, J. Appl. Phys. **89**, 5209 (2001).
[2]K. Sin, S. Funada, M. R. Gibbons, W. Jensen, C. Hiner, X. Shi, and H.-C. Tong, Intermag Digests of Technical Papers, 2002 (unpublished), ER-10; http://ieeexplore. ieee.org/iel5/7849/21611/01001185.pdf
[3]J. Wolfman, D. Mauri, T. Lin, J. Yang, and T. Chen, J. Appl. Phys. **97**, 123713 (2005).
[4]W. F. Egelhoff, Jr., P. J. Chen, C. J. Powell, M. D. Stiles, R. D. McMichael, J. H. Judy, K. Takano, and A. E. Berkowitz, J. Appl. Phys. **82**, 6142 (1997).
[5]B. D. Schrag, A. Anguelouch, S. Invarsson, G. Xiao, Y. Lu, P. L. Trouilloud, A. Gupta, R. A. Wanner, W. J. Gallagher, P. M. Rice, and S. S. P. Parkin, Appl. Phys. Lett. **77**, 2373 (2000).
[6]W. F. Egelhoff, Jr., L. Gan, P. J. Chen, C. J. Powell, R. D. McMichael, R. A. Fry, G. Beach, D. Martien, and A. E. Berkowitz, Mater. Res. Soc. Symp. Proc. **674**, T1.2.1 (2001).
[7]P. H. Holloway and J. B. Hudson, Surf. Sci. **43**, 123 (1974);

S. Hildebrandt, C. Hagendorf, T. Doege, C. Jeckstiess, R. Kulla, H. Neddermeyer, and T. Uttich, J. Vac. Sci. Technol. A **18**, 1010 (2000).
[8]J. D. R. Buchanan, T. P. A. Hase, B. K. Tanner, P. J. Chen, L. Gan, C. J. Powell, and W. F. Egelhoff, Jr., Phys. Rev. B **66**, 104427 (2002); J. D. R. Buchanan, T. P. A. Hase, B. K. Tanner, P. J. Chen, L. Gan, C. J. Powell, and W. F. Egelhoff, Jr., J. Appl. Phys. **93**, 8044 (2003); J. D. R. Buchanan, T. P. A. Hase, B. K. Tanner, C. J. Powell, and W. F. Egelhoff, Jr., *ibid.* **96**, 7278 (2004).