Published in final edited form as:

J Appl Phys. 2017 November ; 122(18): . doi:10.1063/1.4996497.

Reduced resistance drift in tunnel junctions using confined tunnel barriers

Z. S. Barcikowski^{1,2} and J. M. Pomeroy²

¹Department of Materials Science and Engineering, University of Maryland, College Park, Maryland 20740, USA

²National Institute of Standards and Technology, Gaithersburg, Maryland 20899-8423, USA

Abstract

Metal-insulator-metal (MIM) tunnel junctions with the aluminum oxide tunnel barriers confined between cobalt electrodes exhibit less resistance drift over time than junctions that utilize a thick, unconfined aluminum electrode. The improved long time stability is attributed to better initial oxide quality achieved through confinement (use of a potential energy well for the oxygen) and plasma oxidation. In this work, Co/AlO_x/Co and Co/Al/AlO_x/Co tunnel junction aging is compared over a period of approximately 9 months using transport measurements and Wentzel-Kramers-Brillouin (WKB) based modelling. The Co/AlO_x/Co (confined) tunnel junction resistance increased by (32 ± 6) % over 5400 h, while Co/Al/AlO_x/Co (unconfined) tunnel junction resistance increased by (85 ± 23) % over 5200 h. Fit parameters for the tunnel barrier width and potential energy barriers were extracted using WKB transport modelling. These values change only a small amount in the confined Co/AlO_x/Co tunnel junction but show a significant drift in the unconfined Co/AlO_x/Co tunnel junction.

I. INTRODUCTION

Ultrathin aluminum oxide (AlO_x) layers are a popular choice as a tunnel barrier in metal insulator metal (MIM) tunnel junctions implemented for Josephson junction devices, magnetic storage, and in spintronics applications.^{2–7} This popularity is driven largely by the ease of fabricating ultrathin AlO_x tunnel barriers and the essentially uniform attenuation of different spin and symmetry states. For example, spin dependent tunneling experiments utilizing AlO_x tunnel barriers show spin conservation across AlO_x.⁸ However, an undesirable property of AlO_x is “aging” as manifest in drifting electrical properties over time.^{9–12}

The physical changes in the barrier that are associated with resistance drift include diverse phenomena, such as diffusion of electrode/barrier cationic species, oxygen diffusion, glassy relaxation, hydroxyl dissociation, out-diffusion of hydrogen from barrier electrodes, and adsorption of foreign species.^{7,9,10,13–16} Unintentional oxidation of the electrodes adjacent to the tunnel barrier can also cause drastic changes in RA (resistance-area) product and the tunneling magnetoresistance (TMR) ratio.³ The time dependent chemical and structural relaxation of the oxygen profile changes the potential energy landscape in the device, resulting in drifting (aging) of the tunnel junction resistance. Successful techniques to

suppress this aging include surface nitridation, substrate cleaning, and annealing.^{7,10,14} Surface nitridation mitigates diffusion of ambient species but does not address diffusion of species between the tunnel barrier and adjacent (buried) layers. Annealing hastens the glassy relaxation of the tunnel barrier by allowing atoms to (re)arrange into lower free energy configurations, but still may result in a large absolute change from the unannealed resistance and may also produce undesirable diffusion elsewhere in the tunnel junctions.

Here, we report on the fabrication and measurement of a set of representative tunnel junction devices that use cobalt layers on either side of an AlO_x tunnel barrier layer that help “confine” the oxygen to within the designed tunnel barrier. Cobalt is less likely to bond with oxygen than to the aluminum because cobalt oxides, CoO ($\Delta G_f^0 = -215 \text{ kJ/mol}$) and Co₃O₄ ($\Delta G_f^0 = -770 \text{ kJ/mol}$), have much less negative (smaller) free energies of formation than aluminum oxide, Al₂O₃ ($\Delta G_f^0 = -1581 \text{ kJ/mol}$).^{4,17} Therefore, the aluminum layer can be considered a potential well for oxygen. In general, metals whose oxides have relatively small (less negative) standard Gibbs free energy of formation (ΔG_f^0) compared to Al are expected to provide a qualitatively similar benefit. In our experiment, Co thin films are deposited directly before and after AlO_x fabrication to “confine” the oxygen in the tunnel barrier.

(ΔG_f^0 values are taken from Ref. 4.) In this paper, we argue that the smaller free energy of Co oxide can therefore sharpen the oxygen density profile at the interface between the tunnel barrier and the underlying Co layer, such that more oxygen stays within the Al achieving a more uniform Al₂O₃ stoichiometry compared to no confinement. The Co layer deposited after oxidation is then intended to help confine oxygen adsorbed on the AlO_x within the barrier as well. Aging (resistance drift) is typically worse in under-oxidized barriers (glassy relaxation, diffusion of oxygen into the barrier, absorption of foreign species, etc.) and is mitigated by higher oxygen concentrations within the barrier. Our confinement strategy is another method to increase the as-formed oxygen concentration.^{11,14,18,19}

In this paper, confinement using cobalt results in substantially less resistance drift in time. Both the confined and unconfined representative devices reported here also utilize plasma oxidation, which produces oxides with relatively high oxygen concentration (i.e., relatively higher specific resistances) in shorter fabrication times when compared to thermal oxidation,^{3,8,20–22} but additional benefit is seen from the confinement. The stability and aging of the junctions is monitored by current-voltage (I–V) measurements taken periodically over several months. The effective tunnel barrier width x and top/forward (bottom/backward) potential energy height ϕ_t (ϕ_b) are extracted using a WKB (Wentzel-Kramers-Brillouin) transport model for both confined and unconfined tunnel junctions using the approach from Chow.¹

II. EXPERIMENT

The tunnel junctions studied here are fabricated in a custom multi-chamber ultra-high vacuum (UHV) system with base pressure $<10^{-7} \text{ Pa}$ (10^{-9} Torr) modeled after that discussed in Ref. 23. Throughout the entirety of the processing, the wafer is held in a stainless-steel cassette and in electrical contact with the chamber ground. The sample is nominally at room

temperature throughout the fabrication process and the thermocouple on the manipulator shows <3 °C heating. All metallic layers, except for the maskless layers described in steps 2 and 3 below, are deposited through etched Ni shadow masks held ≈ 1.5 mm in front of the wafer onto 75 mm thermally oxidized Si wafers using electron beam deposition. No photolithography, photolithographic resists, developers, etc., are used at any time during fabrication. The wafer is held face down at normal incidence to the electron beam crucible, located ≈ 46 cm. Furthermore, the electron beam crucibles move in-line such that each material's crucible is always located in the same position directly beneath the wafer. Oxidation is done via an oxygen plasma. During plasma oxidation, Al follows a Mott-Cabrera growth behavior, where the oxide thickness scales logarithmically with time.²⁴ The plasma enhances the field-assisted diffusion of Al to the oxidation surface. Shown in Fig. 1(a) is a micrograph of a finished die with two crossed wire tunnel junctions and the corresponding layer structures described below. Tunnel junction areas are determined *post facto* from *ex situ* microscope measurements, which for the devices highlighted here are $3700 \mu\text{m}^2$ (confined) and $3200 \mu\text{m}^2$ (unconfined). After cleaning the wafers *in situ* with an oxygen plasma, the steps for fabricating devices are as follows: (1) a "bottom" mask is in place while depositing 40 nm of Co at ≈ 0.05 nm/s. (2) For the confined devices only, 1 nm of Co is then deposited without a mask to improve adhesion and structural uniformity of Al on the sidewalls and transition region to the SiO_x surface. (3) Then, for the confined devices, 1.1 nm of Al is deposited on the entire wafer without a mask at ≈ 0.02 nm/s, while for the unconfined tunnel junctions, 10 nm Al is deposited through the "bottom" mask. (4) The wafer is transferred *in situ* to an oxygen plasma chamber and oxidized with 21 Pa (160 mTorr) of research grade oxygen using 57 W to 60 W (100 mA at -570 V to -600 V) DC for 7 s. The wafer is held flush and electrically contacted with a grounded circular anode plate by a wafer elevator. The cathode plate is positioned concentrically 4 cm above the wafer in a parallel plate geometry. The cathode and anode plates are stainless steel. X-ray photoelectron spectroscopy performed *in-situ* after oxidation shows no signs of contaminants from sputtering such as Fe, C, Cr, etc., and minimal or no formation of Co oxides. (5) After oxidation, the wafer is held under vacuum for at least 12 h to allow the oxide to relax—in this study, the confined (unconfined) wafer sat for 24 h (21 h). Empirical observations in our group have shown that deposition of a top electrode directly after oxidation results in much higher rates of initial resistance drift (not shown). (6) The wafer is moved back to the deposition chamber *in situ* where a "top" mask is used while depositing 5 Co/40 Cu/5 Au (all in nm) at ≈ 0.05 nm/s for the capping layers. At no time during fabrication is the device removed from vacuum or exposed to pressures above 10^{-6} Pa (10^{-8} Torr) (except for research grade oxygen during plasma oxidation). A cartoon of the completed layer structure in the area where the two wires overlap is shown in Fig. 1(b). In the 10 nm Al case, the oxygen plasma forms a thin oxide skin on top of the thicker Al film where the lower AlO_x edge is not confined by Co and probably not abrupt. After removal from vacuum, all samples are stored in a nitrogen dry-box environment when not under measurement.

Several device wafers with crossed-wire tunnel junctions were processed using the confined architecture and all measurable devices (≈ 10) for this study had resistance-area (RA) products in the range of $(2.6 \text{ to } 3.4) \times 10^4 \Omega \cdot \mu\text{m}$. Device yield was dominated by proper in-

situ alignment of the subsequent shadow masks. The nominal resistance values are obtained using a four-point measurement with 1 mA current for each measurement ($V-I-$ on the bottom electrode and $V+I+$ on the top electrode). The resistance between disconnected electrodes is measured on every wafer to ensure that devices are well defined and free of current shunting. Previous transmission electron microscopy (TEM) measurements show continuous and conformal tunnel barrier formation.²⁵ Furthermore, the device resistance as a function of temperature increases $\approx 20\%$ with decreasing temperature (300 K to 4 K) and fits a $G \propto T^2$ relationship well, supporting our interpretation of high quality barriers with minimal pinhole formation.^{26,27} The resistance values shown are corrected for a “negative resistance” artifact known to occur in crossed wire geometries.²⁸ The initial RA products of these devices are $(3.1 \pm 0.7) \times 10^4 \Omega \cdot \mu\text{m}^2$ for confined and $(7.3 \pm 2.2) \times 10^4 \Omega \cdot \mu\text{m}^2$ for unconfined. Estimated uncertainties derive from the corrected device resistance (due to negative resistance)²⁸ and tunnel junction area measurements.²⁹ Data shown here are taken from devices in the same position on their respective wafers to minimize systematic variations. The device resistances were subsequently measured every 1 to 2 weeks for approximately 8 months. As seen in the figure, the confined devices show significantly less resistance change over the first 1500 h, after which the unconfined device ages at a similar rate as the confined device. Over the length of the study, the confined junction resistance increased by $(32.4 \pm 5.8)\%$ and the unconfined device increased by $(85.1 \pm 23.0)\%$. As a note, the devices presented here are representative samples from a very large number of devices made using the same processes spanning over a decade.^{23,30,31}

To determine the effective thickness and potential barrier heights of the tunnel barriers, we analyzed $\frac{dI}{dV}(V)$ curves for each type of device as shown in Fig. 2(b). The $\frac{dI}{dV}(V)$ data are obtained from numerical differences of $I-V$ data. The fits are done within a bias window of ± 100 mV in order to stay in the low voltage regime (compared to the barrier height).^{1,29} All conductance curves are approximately parabolic, signifying tunneling as the dominant transport mechanism. Like the results of the single point resistance measurements, $\frac{dI}{dV}(V)$ data of the confined devices show little change [all $\frac{dI}{dV}(V)$ are overlapping] during the course of the study, while the unconfined devices change significantly over time.

To fit the $\frac{dI}{dV}(V)$ measurements and extract the effective barrier parameters, we use a WKB transport model summarized below, following the approach of Chow.¹ For two metallic electrodes with uniform and structure-less (free electron gas) densities of states separated by a thin insulating barrier, the simplified equation for tunneling current density is given by

$$J(V) = \frac{4\pi me}{h^3} \int_0^{E_m} \rho(E_x, V) D(E_x) dE_x, \quad (1)$$

where E_m is the maximum energy of electrons in the electrodes, m is the free electron mass, e is the elementary charge, h is Planck's constant, E_x is the energy of an electron associated with momentum in the tunneling direction, V is the applied voltage, $D(E_x)$ is the tunneling

probability, and $\rho(E_x, V)$ is the supply function which includes the Fermi-Dirac distributions and densities of states of the electrodes. Within the WKB approximation, the tunneling probability is

$$D(E_x) = \exp \left\{ -\frac{4\pi(2m)^{\frac{1}{2}}}{h} \left(\int_{x_1}^{x_2} [\phi(x) + E_F - E_x]^{\frac{1}{2}} dx \right) \right\}. \quad (2)$$

In this equation, x_1 and x_2 represent the (effective) beginning and end of the potential barrier, $\phi(x)$ is the potential barrier relative to E_F , and E_F is the Fermi level of the negatively biased (reference) electrode. To solve the integral in Eq. (2), $\phi(x)$ is modeled as a trapezoidal barrier with effective barrier width $x = x_2 - x_1$ and barrier height ϕ_r , given in Eq. (3), and illustrated in the potential energy diagram shown in Fig. 3. This approximation of an arbitrary barrier is numerically equivalent to that used by Simmons as shown in the Appendix of Ref. 32

$$\phi_r = \left\{ \frac{1}{\Delta x} \int_{x_1}^{x_2} [\phi(x)]^{\frac{1}{2}} dx \right\}^2 = \frac{4}{9} \left(\frac{\phi_b^{\frac{3}{2}} - (\phi_t - V)^{\frac{3}{2}}}{\phi_b - \phi_t + V} \right)^2. \quad (3)$$

On the right side of Eq. (3), ϕ_t is the “top” barrier height and ϕ_b is the “bottom” barrier height [i.e., left and right barrier heights as seen in Fig. 3(a)]. This approximation is sensitive to an asymmetric barrier, i.e., $\phi_t \neq \phi_b$, which can arise from work function differences or unequal oxidation states. Substitution of Eq. (3) into Eq. (2) results in the tunneling probability given by the following equation:

$$D(E_x) = \exp \left\{ -\frac{4\pi(2m)^{\frac{1}{2}}}{h} \Delta x (\phi_r + E_F + E_x)^{\frac{1}{2}} \right\}. \quad (4)$$

After substitution of Eq. (4) into Eq. (1), the integration can be done using the metallic densities of states and the Fermi-Dirac distribution within the supply function. The tunneling current density is then given by

$$J(V) = \frac{3e(2m)^{\frac{1}{2}}}{\beta(h\Delta x)^2} \left\{ \phi_r \exp \left[-\beta \Delta x \phi_r^{\frac{1}{2}} \right] - (\phi_r + eV) \times \exp \left[-\beta \Delta x (\phi_r + eV)^{\frac{1}{2}} \right] \right\}, \quad (5)$$

where $\beta = \frac{4\pi(2m)^{\frac{1}{2}}}{h} \approx 1.025 \text{ eV}^{-\frac{1}{2}} \cdot \text{\AA}^{-1}$.

The equation for conductance $G(V)$ is arrived at by multiplying the device area A by the

derivative of Eq. (5) with respect to V , or $G(V) = A g(V) = A \frac{dJ(V)}{dV}$. Shown in Fig. 3(b) are

conductance data for a confined tunnel junction device [Day 214 in 2(B)] with a fit using the transport model described earlier, where ϕ_b , ϕ_t , and x are the free parameters in the fit. Filled squares are the raw data, and the solid line is the transport model fit, showing good agreement with the data. All the effective barrier parameters discussed below are obtained by fitting using the above model. Previous TEM measurements have shown our barriers to be sufficiently flat and parallel within the context of this model.²⁵ However, the extracted fit parameters should be considered “effective” since the exact values depend on choices of scaling parameters like the electron effective mass.

Shown in Table I are the initial (nominal $t = 0$) barrier parameters extracted from the fits. The initial 1.1 nm of elemental Al deposited is expected to expand by approximately $\approx 28\%$ when oxidized to be Al_2O_3 , resulting in a physical barrier width of 1.4 nm.³ The difference between 1.4 nm and the fitted values of the confined barrier width of $1.28 \text{ nm} \pm 0.06 \text{ nm}$ is within the uncertainty in the initial Al layer thickness, the difference in electrical and physical widths due to effects like the image charge potentials (which tend to electrostatically round off the barrier), and non-uniformities across the device area.³² The forward and backward barrier heights for the confined are the same within the uncertainties, $\phi_t = 1.10 \text{ eV} \pm 0.06 \text{ eV}$ and $\phi_b = 1.13 \text{ eV} \pm 0.06 \text{ eV}$, indicating that the barrier is uniformly oxidized. These barrier heights and width values show good agreement with the literature for similar Co/AlOx/Co tunnel junctions.^{17,33,34}

In contrast, the fit values for the unconfined device show a relatively large effective barrier width of $2.03 \text{ nm} \pm 0.10 \text{ nm}$ despite using the same oxidation parameters as the confined device. Furthermore, the barrier height fit parameters of the unconfined device are less than half of the values of the confined device with $\phi_t = 0.46 \text{ eV} \pm 0.02 \text{ eV}$ and $\phi_b = 0.41 \text{ eV} \pm 0.02 \text{ eV}$. This finding suggests that the unconfined tunnel barrier is not as thoroughly oxidized as the confined tunnel barrier, as discussed below. Additionally, the finding of a larger effective thickness in the unconfined device is consistent with the larger initial specific resistance, due to the inverse square dependence of the current density on the barrier thickness in the prefactor of Eq. (5). The specific resistance difference is understood using only tunneling transport.

In addition to the resistance, we monitored how the best fits for the barrier parameters changed throughout this study, providing insight into the mechanisms of the resistance drift. Figures 4(a) and 4(b) show the normalized fit values for barrier width and barrier heights over time, respectively. For the confined device, the barrier width, x , shows very little change over 5400 h. The barrier heights of the confined device also show a small upward drift, as ϕ_t increases by $(13 \pm 1)\%$ and ϕ_b increases by $(4 \pm 2)\%$. The unconfined device shows substantially more drift in all three parameters. Specifically, the effective barrier width, x , decreases by $(9.9 \pm 0.1)\%$ over 5200 h, while ϕ_t increases by $(35 \pm 3.5)\%$ and ϕ_b increases by $(45 \pm 4.5)\%$.

III. RESULTS AND DISCUSSION

Resistance drift of the kind observed in this work is understood to be associated with an evolution of the oxygen concentration profile in the tunnel barrier. Aluminum oxides that

have sub-stoichiometric and/or nonuniform oxygen concentration profiles have a large phase space of atomic configurations and undergo a long process of relaxation. As the atomic configurations change, the local electronic states change, resulting in the electrical resistance drift.¹¹ Typically, during the initial fabrication of AlO_x , the aluminum closest to the surface is oxygen-rich compared to deeper in the aluminum.^{8,35} Once the oxidation is complete, the oxygen concentration profile relaxes with time and temperature from this asymmetric AlO_x towards a more uniform Al_2O_3 -like stoichiometry throughout the barrier.³⁵ Annealing the barriers accelerates this relaxation process, helping the material to reach a lower energy configuration.¹⁰ Alternatively, improving the oxygen concentration profile created during the initial oxidation can also reduce the resistance drift, since the system is initially closer to a relaxed state.^{36,37} It should be noted that the use of plasma oxidation plays an important role in this context, as oxides formed by plasma oxidation opposed to thermal oxidation typically have much higher O:Al ratios which are closer to stoichiometric Al_2O_3 .⁸ While we have only studied plasma oxides, the use of confinement is expected to benefit oxide tunnel barriers independent of oxidation method. As another approach, some groups have formed the barrier by sequential partial depositions and oxidations to improve the uniformity of the tunnel barriers and reduce the resistance drift.³⁵

For the work reported here, we can exclude several causes of aging identified in other studies, e.g., (1) aging induced by photoresist residue is not applicable since photolithography was not used here; (2) aging induced by disassociation of hydroxyls in the barrier is unlikely as junctions described here were not exposed to elevated temperatures.¹³ Also, diffusion of oxygen and other foreign species from air is not likely to have a significant impact on aging since the tunnel barriers are deeply buried beneath the overgrown electrodes and the tunnel junction areas are large relative to the diffusion lengths from sidewalls.^{7,13} Furthermore, we do not expect significant out-diffusion of hydrogen from the electrodes to the tunnel barrier given the reported room temperature values for hydrogen diffusivity in cobalt.³⁸

To understand the resistance drift, and its suppression in the confined case, we first note that plasma oxidation is used to form the barrier. A sub-stoichiometric Al-rich oxidation front moves through the metallic Al, with oxide forming initially at Al grain boundaries. The O:Al ratio does not increase significantly until the oxidation front reaches an underlying layer acting as a diffusion stop, e.g., the cobalt under layer in our tunnel junctions.^{8,36} Compared to the confined case, the unconfined barrier does not have an oxidation stop beneath the barrier and the WKB fit parameters confirm a relatively wide barrier with low barrier heights (less oxygen), which contracts with increasing time. In the confined devices, the large difference in ΔG_f^0 between Al_2O_3 and the Co oxides allows all of the metallic Al to be oxidized prior to the oxidation front moving beyond the Al.^{3,4,24,35} Thus, our results can potentially be explained by a relatively sharper and more homogenous oxygen concentration profile within the tunnel barrier for the confined case due to this potential well for oxygen. We acknowledge that high resolution TEM (HRTEM) studies find sharp, atomic interfaces at the oxide/metal interface in similar unconfined devices, but the tunnel barrier properties were observed to vary between different measurement locations and no statements are made regarding the interpreted oxygen profiles.^{20–22} The 1.3 nm barrier width and 1.1 eV barrier

heights from the WKB fits of the confined barrier support the conclusion that the thin Al is well oxidized with an oxygen concentration much closer to that of Al_2O_3 compared to the unconfined tunnel junction devices.^{8,24,33} Consequently, the aging is less severe, due to the initial oxygen profile being more homogeneous and the stoichiometry closer to that of Al_2O_3 .^{11,19,37}

IV. CONCLUSION

In conclusion, using free energy confinement in combination with plasma oxidation for AlOx tunnel barriers results in much less aggressive resistance drift over time (“aging”) than without confinement. Tunnel barrier parameters extracted using a WKB transport model support the interpretation that Co confinement acts to confine the oxygen, resulting in an ultrathin barrier with high potential energy barrier heights compared to the unconfined case. Over time, the confined tunnel junction barrier width, x , and barrier heights, parameters ϕ_t and ϕ_b extracted from the WKB fits, also remain stable. The improved initial barrier stoichiometry and homogeneity in the confined tunnel junction device is likely responsible for less aggressive aging. Furthermore, these results are consistent with other approaches designed to result in a thoroughly oxidized barrier, such as utilizing diffusion stops and sequential deposition and oxidations, which mitigate aging without additional thermal budget that can be potentially deleterious during annealing.

Acknowledgments

The authors acknowledge Yanxue Hong, Dr. Michael Stewart, and Dr. Neil Zimmerman at the National Institute of Standards and Technology (NIST) for helpful discussion and comments. Device fabrication techniques and laboratory best practices were derived from Dr. Russell Lake’s past work at NIST. Profilometry measurements were performed at the Center for Nanoscale Science and Technology (CNST) facility at NIST.

References

1. Chow CK. J. Appl. Phys. 1965; 36(2):559.
2. Konkin MK, Adler JG. J. Appl. Phys. 1979; 50(12):8125.
3. Kuiper AET, Gillies MF, Kottler V, ‘t Hooft GW, van Berkum JGM, van der Marel C, Tamminga Y, Snijders JHM. J. Appl. Phys. 2001; 89(3)
4. LeClair P, Kohlhepp JT, Smits AA, Swagten HJM, Koopmans B, De Jonge WJM. J. Appl. Phys. 2000; 87(9):6070–6072.
5. Morohashi SI, Yoshida A, Hasuo S. J. Appl. Phys. 1991; 70(3):1806.
6. Scherer H, Weimann T, Zorin AB, Niemeyer J. J. Appl. Phys. 2001; 90(5):2528.
7. Shiota T, Imamura T, Hasuo S. IEEE Trans. Appl. Supercond. 1992; 2(4)
8. Knechten CAM. Eindhoven University of Technology. 2004
9. Dochev D, Pavolotsky A, Lai Z, Belitsky V. J. Phys.: Conf. Ser. 2010; 234(4):042006.
10. Koppinen PJ, Väistö LM, Maasilta IJ. Appl. Phys. Lett. 2007; 90(5):053503.
11. Nesbitt JR, Hebard AF. Phys. Rev. B. 2007; 75(19)
12. Pavolotsky AB, Dochev D, Belitsky V. J. Appl. Phys. 2011; 109(2):024502.
13. Gates JV, Washington MA, Gurvitch M. J. Appl. Phys. 1984; 55(5):1419.
14. Pop IM, Fournier T, Crozes T, Lecocq F, Matei I, Pannetier B, Buisson O, Guichard W. J. Vac. Sci. Technol. B: Microelectron. Nanometer Struct. 2012; 30(1):010607.
15. Sousa RC, Sun JJ, Soares V, Freitas PP, Kling A, da Silva MF, Soares JC. Appl. Phys. Lett. 1998; 73(22):3288.

16. Tolpygo SK, Amparo D. *Supercond. Sci. Technol.* 2010; 23(3):034024.
17. Lee D, Hong J. *J. Appl. Phys.* 2005; 97(9):093905.
18. Batlle X, Cuadra PJ, Zhang Z, Cardoso S, Freitas PP. *J. Magn. Magn. Mater.* 2003; 261(3):L305–L310.
19. DuBois TC, Cyster MJ, Opletal G, Russo SP, Cole JH. *Mol. Simul.* 2015;1–7.
20. Aghdam PR, Rashid HM, Pavolotsky A, Desmaris V, Belitsky V. *J. Appl. Phys.* 2016; 119:054502.
21. Zeng LJ, Nik S, Greibe T, Krantz P, Wilson CM, Delsing P, Olsson E. *J. Phys. D: Appl. Phys.* 2015; 48(39):395308.
22. Aref T, Averin A, van Dijken S, Ferring A, Koberidze M, Maisi VF, Nguyend HQ, Nieminen RM, Pekola JP, Yao LD. *J. Appl. Phys.* 2014; 116(7):073702.
23. Lake RE, Pomeroy JM, Grube H, Sosolik CE. *Phys. Rev. Lett.* 2011; 107(6):063202. [PubMed: 21902321]
24. Gillies MF, Kuiper AET, Coehoorn R, Donkers JJTM. *J. Appl. Phys.* 2000; 88(1)
25. Pomeroy JM, Grube H, Sun PL, Lake RE. *AIP Conf. Proc.* 2010; 13:111–114.
26. Lai Y-R, Yu K-F, Lin Y-H, Wu J-C, Lin J-J. *AIP Adv.* 2012; 2(3):032155.
27. Rüdiger U, Calarco R, May U, Samm K, Hauch J, Kittur H, Sperlich M, Güntherodt G. *J. Appl. Phys.* 2001; 89(11):7573–7575.
28. Pomeroy JM, Grube H. *J. Appl. Phys.* 2009; 105(9):094503–094508.
29. Lake R. *Clemson University.* 2012
30. Pomeroy JM, Lake RE. *Nucl. Instrum. Methods Phys. Res., Sect. B.* 2013; 317:66–71.
31. Pomeroy JM, Grube H, Perrella AC, Gillaspay JD. *Appl. Phys. Lett.* 2007; 91(7):073506.
32. Simmons JG. *J. Appl. Phys.* 1963; 34(6):1793.
33. Arakawa N, Otaka Y, Shiiki K. *Thin Solid Films.* 2006; 505(1–2):67–70.
34. Kurnosikov O, de Nooij FC, LeClair P, Kohlhepp JT, Koopmans B, Swagten HJM, de Jonge WJM. *Phys. Rev. B.* 2001; 64(15)
35. Holmqvist T, Meschke M, Pekola JP. *J. Vac. Sci. Technol. B: Microelectron. Nanometer Struct.* 2008; 26(1):28.
36. Yoon KS, Park JH, Choi JH, Yang JY, Lee CH, Kim CO, Hong JP, Kang TW. *Appl. Phys. Lett.* 2001; 79(8):1160.
37. Zimmerman NM, Huber WH, Simonds B, Hourdakis E, Fujiwara A, Ono Y, Takahashi Y, Inokawa H, Furlan M, Keller MW. *J. Appl. Phys.* 2008; 104(3):033710.
38. Caskey GR Jr, Derrick RG, Louthan MR Jr. *Scr. Metall.* 1974; 8:481–486.

FIG. 1.

(a) Photograph of die “B1” from a finished 75 mm wafer. The inset is the optical microscope image of “B” geometry, i.e., a crossed wire device. (b) Schematic cross section of the tunnel junction at the intersection of crossed wires in (a). Aluminum films are deposited on the cobalt bottom electrode and plasma oxidized. Aluminum films are 1.1 nm for confined devices and 10 nm for unconfined devices, both oxidized under the same parameters.

FIG. 2.

(a) Normalized resistance drift over time for the confined Co/AlOx/Co (unfilled squares) and unconfined Co/Al/AlOx/Co (filled squares) tunnel junctions measured in a four-point geometry (initial R_0 values shown in the legend). (b) Normalized differential conductance curves for confined (unfilled) and unconfined (filled) tunnel junctions. Three curves each taken ≈ 100 days apart are displayed to show the relative amount of change seen in the unconfined devices compared to the confined devices, which are relatively indistinguishable as they are all overlapping.

FIG. 3.

(a) Potential energy diagram for an asymmetric tunnel barrier with a small positive bias on electrode 1 (left) as modeled in the WKB approximation.¹ (b) Differential conductance data from the confined tunnel junction on day 1 after fabrication (symbols) fitted with the WKB model (line). The fit parameters extracted for this curve are $x = 1.3 \text{ nm} \pm 5.0 \%$ barrier width, $\phi_t = 1.2 \text{ eV} \pm 5.0 \%$, and $\phi_b = 1.2 \text{ eV} \pm 5.0 \%$.

(a)

(a)

FIG. 4.

(a) Effective tunnel barrier width, Δx , over time. (B) Barrier heights, $\phi_{t,b}$, for confined Co/AlOx/Co (unfilled) and unconfined Co/Al/AlOx/Co (filled) tunnel junctions. Both ϕ_t and ϕ_b are shown. Barrier width and barrier height values extracted using the WKB transport model discussed in the text.¹

TABLE I

Day 1 best fit parameters effective barrier width, x , and effective barrier heights, ϕ_t and ϕ_b , extracted using the WKB transport model from measurements of the confined (Co/AlOx/Co) and unconfined (Co/Al/AlOx/Co) tunnel junction devices.

	Confined	Unconfined
ϕ_t (eV)	1.10 ± 0.06	0.46 ± 0.02
ϕ_b (eV)	1.13 ± 0.06	0.41 ± 0.02
x (nm)	1.28 ± 0.06	2.03 ± 0.10