
This is an electronic reprint of the original article.
This reprint may differ from the original in pagination and typographic detail.

Holmqvist, Tommy; Meschke, Matthias; Pekola, Jukka
Double oxidation scheme for tunnel junction fabrication

Published in:
Journal of Vacuum Science and Technology. Part B.

DOI:
[10.1116/1.2817629](https://doi.org/10.1116/1.2817629)

Published: 01/01/2008

Document Version
Publisher's PDF, also known as Version of record

Please cite the original version:
Holmqvist, T., Meschke, M., & Pekola, J. (2008). Double oxidation scheme for tunnel junction fabrication. *Journal of Vacuum Science and Technology. Part B.*, 26(1), 28-31. <https://doi.org/10.1116/1.2817629>

This material is protected by copyright and other intellectual property rights, and duplication or sale of all or part of any of the repository collections is not permitted, except that material may be duplicated by you for your research use or educational purposes in electronic or print form. You must obtain permission for any other use. Electronic or print copies may not be offered, whether for sale or otherwise to anyone who is not an authorised user.

Double oxidation scheme for tunnel junction fabrication

T. Holmqvist, M. Meschke, and J. P. Pekola

Citation: *Journal of Vacuum Science & Technology B: Microelectronics and Nanometer Structures Processing, Measurement, and Phenomena* **26**, 28 (2008); doi: 10.1116/1.2817629

View online: <http://dx.doi.org/10.1116/1.2817629>

View Table of Contents: <http://avs.scitation.org/toc/jvn/26/1>

Published by the [American Institute of Physics](#)

Double oxidation scheme for tunnel junction fabrication

T. Holmqvist,^{a)} M. Meschke, and J. P. Pekola

*Low Temperature Laboratory, Helsinki University of Technology, P.O. Box 3500,
FIN-02015 TKK, Finland*

(Received 10 August 2007; accepted 31 October 2007; published 2 January 2008)

The authors report a method to achieve Al–AlO_x–Al tunnel junctions with high specific resistance in a controlled manner using a double oxidation technique. The technique is based on the standard method for oxidation repeated on an additional Al layer. The tunnel junctions were characterized with standard methods, such as comparison of room temperature resistance with liquid helium resistance and the authors found them to be of comparable quality to junctions fabricated with standard single oxidation. Fitting with the Simmons model suggests that both the barrier width and barrier height are consistent with those obtained in a single oxidation step. The junction specific capacitance was determined at low temperature to be 68 fF/μm². These junctions, employed in low temperature measurements and applications, demonstrate expected and stable conductance characteristics. The double oxidation method is straightforward to implement in a basic setup for tunnel junction fabrication. © 2008 American Vacuum Society. [DOI: 10.1116/1.2817629]

I. INTRODUCTION

While submicron scale metal-insulator-metal (MIM) tunnel junctions have found much attention and practical applications, there are still unresolved issues with the micron scale junctions as these mature into commercial applications.¹ The most common material for the dielectric layer is thermally oxidized aluminum, AlO_x. The procedure for producing junctions with this barrier is straightforward: deposit clean Al in a vacuum chamber and flow oxygen into the chamber for a predetermined time at a certain pressure, and cover with a second metal layer. Yet, this standard method for producing tunnel junctions, while straightforward and simple, has a few problems. In particular, it can be difficult to achieve high specific resistance of the tunnel junctions, since the oxide forms a diffusion barrier that shields the underlying metal from further oxidation. Hence, a change of resistance of tunnel junctions over time (i.e., aging effects) has been observed for both large area tunnel junctions² and for small ones.^{3,4}

It is believed that the standard process for fabricating AlO_x produces an oxygen rich compound near to the surface. It relaxes to the stoichiometric stable Al₂O₃ only after some time when oxygen has diffused from the surface into the underlying aluminum,⁵ which has been brought forward as a plausible explanation for aging. It was recently reported that annealing of MIM tunnel junctions stabilizes the junctions,³ lending some support to the idea that aging is related to relaxation of AlO_x to Al₂O₃. One method to improve the quality of tunnel junctions is to form the dielectric layer in several steps.⁶ Obviously, this method has the added advantage of making it possible to fabricate tunnel junctions with high resistance simply by increasing the number of layers. This method has been reported to be in use for magnetic tunnel junction fabrication.⁷ In this article, we describe the fabrication of Al–AlO_x–Al tunnel junctions using a double

oxidation method to form a stable dielectric layer. We have investigated the quality of these tunnel junctions using common criteria by determining specific capacitance and resistance, and testing these in low temperature measurements.

II. FABRICATION

In order to increase the tunnel barrier thickness beyond the limits set by the diffusion barrier of the oxide layer,⁵ a procedure was developed with the main idea being to evaporate a thin (<1 nm) layer of Al on top of already oxidized Al, whereafter the oxidation procedure was repeated. The second aluminum layer is sufficiently thin such that it oxidizes completely, thereby increasing the thickness of aluminum oxide without leaving layers of Al buried inside the junction.

One application of such junctions is Coulomb blockade thermometry.⁸ For this purpose, and in order to facilitate easy characterization of the tunnel junctions, we fabricate junction arrays with seven parallel lines of $N=64$ junctions in each line in series (see Fig. 1). Each metal island between the junctions has a large cooling fin to guarantee sufficient electron thermalization even at temperatures as low as 50 mK and below. The tunnel junction area is about 2.2 μm². The fabrication method for the test structure is based on the shadow evaporation method pioneered by Dolan.⁹ Structures are defined using standard electron beam lithography with a two layer resist scheme. The metals are evaporated with an electron gun source in a vacuum chamber with base pressure of less than 1×10^{-7} mbar. The bottom aluminum electrode is evaporated to a thickness of 30 nm at an oblique angle α with respect to perpendicular orientation. The first aluminum layer is oxidized for 10 min at 100 mbars in pure oxygen. After the first evaporation and subsequent oxidation, another aluminum layer with a thickness of 0.6 nm is evaporated and oxidized under the same conditions as the first one. A quartz crystal thickness monitor (Inficon XTC) with a nominal thickness resolution of 0.1 nm is used to observe the evapo-

^{a)}Electronic mail: tommy.holmqvist@l.tl.tkk.fi

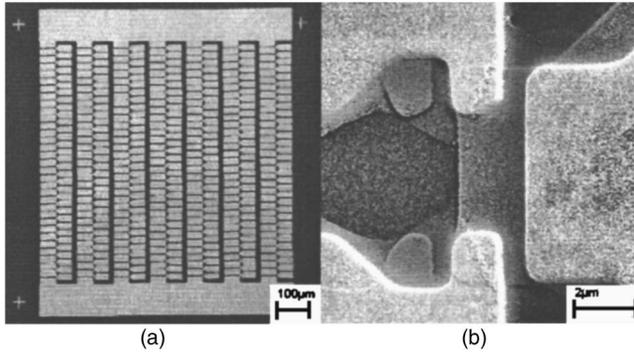


FIG. 1. (a) Scanning electron micrograph of the structure used to measure tunnel junction characteristics. It is an array of seven parallel lines with 64 tunnel junctions in series in each vertical line. Cooling fins are added to the islands between junctions to facilitate low temperature measurements with minimal decoupling of the electrons to the phonon system. (b) Two tunnel junctions of the array with higher magnification. The junction area is nominally $1.5 \times 1.5 \mu\text{m}^2$.

ration rate. When the rate is stable at 2 \AA/s , the shutter is opened for 3 s. The topmost electrode is formed by evaporating aluminum to a thickness of 30 nm at an angle $-\alpha$. The last metal evaporation step is copper to a thickness of 500 nm. With the exception of the thin aluminum layer, deposition rates are kept at $5\text{--}10 \text{ \AA/s}$.

III. QUALITY CONSIDERATIONS

One measure of the quality of a tunnel junction is the change of its resistance when cooled from room temperature to liquid He temperature. It has been suggested that this is a more reliable criterion for pinhole-free junctions than fitting I - V characteristics of the junction with the standard Simmons model.¹⁰ In practice, approximately 15% increase of junction resistance is reported to indicate high quality Al-AIO_x-Al tunnel junctions.¹¹ Since tunnel junction resistance R_j is inversely proportional to the area A of the junction, the junction specific resistance, $R_C = R_j A$, is used as a characteristic figure. Fabrication of large area tunnel junctions with a single oxidation step tends to result in limited specific resistance of the tunnel junction. As seen in Table I, even drastic increases in oxide pressure and oxidation time may result in a relatively moderate increase in junction resistance. The in-

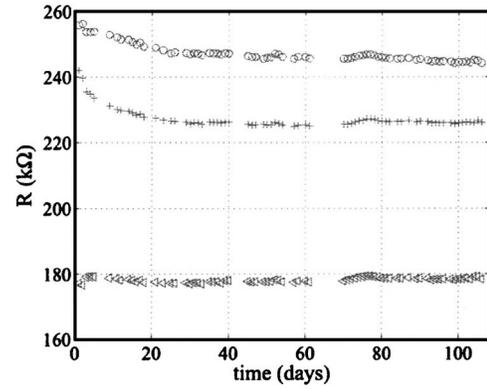


FIG. 2. Resistance of three arrays with tunnel junctions manufactured with double oxidation. All the three samples were fabricated in the same vacuum cycle in batch 3 (see Table I). The resistance was monitored over an extended time while the junctions were stored in ambient air at room temperature. A small decrease of resistances is seen in the first days but then the junctions stabilize. The overall change in resistance is $<10\%$ over this duration.

roduction of a second oxidation step, as seen in batches 3–5 yields consistently specific resistances that are approximately one order of magnitude higher.

One of the more troublesome characteristics of tunnel junctions is aging, which is generally observed as an increase in resistance over time. Recent studies of aging have mainly concerned small area junctions⁴ with one oxidation step. Earlier studies include large area junctions² but, again, only for one oxidation step. In order to monitor aging, three samples fabricated with two oxidation steps from the same batch were monitored over a period of over 100 days while stored under ambient air conditions at room temperature; the result is shown in Fig. 2. It is noteworthy that no increase of the resistance was seen, instead for two of the three studied samples the resistance decreased about 5% during the first few days of the measurement. The change of resistance for any of the three samples was well below 10% over this duration indicating that aging was weak for these junctions.

Fitting bias dependence of conductance to the Simmons model is the standard method to determine both the thickness of the dielectric layer and the height of the potential barrier.¹² Voltage dependence of the differential conductance follows:

TABLE I. Samples fabricated and characterized. Note that the two first batches were fabricated with the standard single oxidation of aluminum. In the table, R_C^{RT} is the characteristic resistance at room temperature and R_C^{LHe} at 4.2 K. ΔR_C is defined as $\Delta R_C = R_C^{\text{LHe}} - R_C^{\text{RT}}$.

Batch	Oxidation pressure (mbar)	Oxidation time (min)	R_C^{RT} ($\text{k}\Omega\mu\text{m}^2$)	R_C^{LHe} ($\text{k}\Omega\mu\text{m}^2$)	$\Delta R_C / R_C^{\text{RT}}$ (%)
1	100	10	3.24	3.78	17
2	200	60	6.14	7.60	24
3	100	10	37.12	45.22	22
4	100	10	33.3	39.82	16
5	100	10	58.3	71.5	23

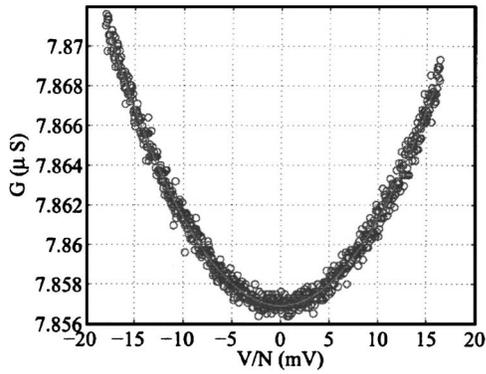


FIG. 3. Measured data (○) of dynamic conductance $G=dI/dV$ plotted vs voltage over one junction. Fitting a parabola (solid line) gives, using the Simmons model, an effective barrier width of $d=12$ Å and a barrier height of $\phi=3$ eV. This measurement was performed at $T=77$ K. The measurement at 4 K gave almost identical results apart from the Coulomb blockade feature around zero bias.

$$dI/dV = G_0(1 + 3\gamma V^2), \quad (1)$$

where G_0 and γ are determined from measurements.^{11,13} Based on conductance measurement shown in Fig. 3, the samples fabricated using double oxidation were found to follow the parabolic dependence of Eq. (1) with a barrier thickness of $d=12$ Å and a barrier height of $\phi=3$ eV which are reasonably in line with values found for tunnel junctions fabricated with one oxidation step only.^{11,14–16} As mentioned in the introduction, a method to verify the quality of the tunnel junctions is to measure the increase of resistance as the junctions are cooled from room temperature to liquid helium temperature. Table I shows that one and two oxidation steps both lead to similar results in this respect.

IV. LOW TEMPERATURE PROPERTIES

In order to determine the exact value of the capacitance of the tunnel junctions, the conductance curve in the partial Coulomb blockade regime was measured at low (~ 50 mK) temperature. Temperature is determined from the half width at full minimum, $V_{1/2}$, of the normalized conductance curve.⁸ The capacitance C of the junctions is determined from the normalized conductance¹⁷

$$C = \left(\frac{N-1}{N} \right) \frac{e^2}{6k_B T} \left(\frac{\Delta G}{G_T} \right)^{-1}. \quad (2)$$

Here, $\Delta G/G_T$ is the normalized depth of the zero bias conductance drop. With values from Fig. 4, Eq. (2) gives $C=152$ fF which, in turn, gives a capacitance per area of 68 fF/ μm^2 . Often cited experimental values for the specific capacitance are values for Nb/ AlO_x /Nb junctions which range between 45 and 60 fF/ μm^2 .^{18,19} Spectroscopic studies indicate that the oxygen content and distribution depends strongly on the work function of the electrodes.²⁰ Therefore the capacitance of a tunnel junction with Nb electrodes may not be comparable to that of a junction formed with Al electrodes. Data for thermally oxidized Al– AlO_x –Al junctions range from slightly below 45 fF/ μm^2 (Refs. 21 and 22) up

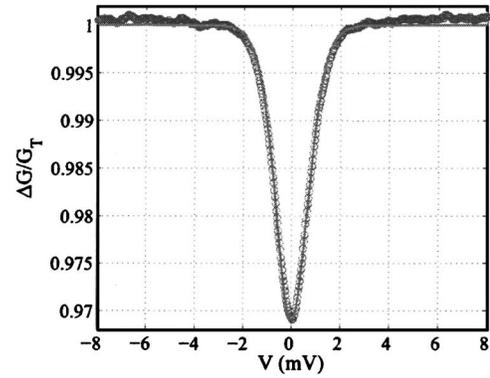


FIG. 4. Measured (○) normalized conductance vs voltage across $N=64$ junctions. Fitting with expression for conductance (solid line) gives together with $eV_{1/2} \approx 5.44Nk_B T$ a temperature of 48.5 mK (Ref. 8). The measurement was carried out in a magnetic field of approximately 50 mT oriented perpendicular to the metal film.

to 220 fF/ μm^2 (Ref. 23) with many falling in the range between 60 and 110 fF/ μm^2 .^{24–27} Our value for specific capacitance is therefore in line with previously measured values for standard Al– AlO_x –Al junctions.

V. CONCLUSIONS

We have made tunnel junctions using a two step oxidation scheme which utilizes standard procedures for MIM tunnel junction fabrication. This scheme results in tunnel junctions with specific resistances in the 30 – 60 k Ω μm^2 range at room temperature, roughly one order of magnitude larger than the typical resistance with a single oxidation step. The capacitance per junction area was found to be about 70 fF/ μm^2 which is similar to those obtained with a single oxidation step. Using accepted criteria, these junctions are found to be of high quality and exhibit only weak aging.

ACKNOWLEDGMENTS

The authors gratefully acknowledge financial support from Centre for Metrology and Accreditation (MIKES) and valuable discussions with A. M. Savin and R. Blaauwgeers.

¹See, e.g., <http://www.hypres.com>

²R. M. Handy, Phys. Rev. **126**, 1968 (1962).

³P. J. Koppinen, L. M. Väistö, and I. J. Maasilta, Appl. Phys. Lett. **90**, 053503 (2007).

⁴See, e.g., K. Bladh, Ph.D. thesis, Chalmers University of Technology, 2005.

⁵L. P. H. Jeurgens, W. G. Sloof, F. D. Tichelaar, and E. J. Mittemeijer, J. Appl. Phys. **92**, 1649 (2002).

⁶J. B. Hansen (private communication).

⁷E. Y. Chen, R. Whig, J. M. Slaughter, D. Cronk, J. Goggin, G. Steiner, and S. Tehrani, J. Appl. Phys. **87**, 6061 (2000).

⁸J. P. Pekola, K. P. Hirvi, J. P. Kauppinen, and M. A. Paalanen, Phys. Rev. Lett. **73**, 2903 (1994).

⁹G. J. Dolan, Appl. Phys. Lett. **31**, 337 (1977).

¹⁰B. J. Jönsson-Åkerman, R. Escudero, C. Leighton, S. Kim, and I. K. Schuller, Appl. Phys. Lett. **77**, 1870 (2000).

¹¹K. Gloos, P. J. Koppinen, and J. P. Pekola, J. Phys.: Condens. Matter **15**, 1733 (2003).

¹²J. G. Simmons, J. Appl. Phys. **34**, 238 (1963).

¹³R. Meservey, P. M. Tedrow, and J. S. Brooks, J. Appl. Phys. **53**, 1563 (1981).

- ¹⁴D. McBride, G. Rochlin, and P. Hansma, *J. Appl. Phys.* **45**, 2305 (1974).
- ¹⁵K. H. Gundlach and J. Hölzl, *Surf. Sci.* **27**, 125 (1971).
- ¹⁶J. Kadlec and K. H. Gundlach, *Solid State Commun.* **16**, 621 (1975).
- ¹⁷S. Farhangfar, K. P. Hirvi, J. P. Kauppinen, J. P. Pekola, J. J. Toppari, D. V. Averin, and A. N. Korotkov, *J. Low Temp. Phys.* **108**, 191 (1997).
- ¹⁸M. Gurvitch, M. A. Washington, and H. A. Huggins, *Appl. Phys. Lett.* **42**, 472 (1983).
- ¹⁹A. W. Lichtenberger, C. P. McClay, R. J. Mattauch, M. J. Feldman, S.-K. Pan, and A. R. Kerr, *IEEE Trans. Magn.* **25**, 1247 (1989).
- ²⁰E. Tan, P. G. Manther, A. C. Perella, J. C. Read, and R. A. Buhrman, *Phys. Rev. B* **71**, 161401 (2005).
- ²¹F. Balestro, J. Claudon, J. P. Pekola, and O. Buisson, *Phys. Rev. Lett.* **91**, 158301 (2003).
- ²²P. D. Dresselhaus, L. Ji, S. Han, J. E. Lukens, and K. K. Likharev, *Phys. Rev. Lett.* **72**, 3226 (1994).
- ²³T. A. Fulton and G. J. Dolan, *Phys. Rev. Lett.* **59**, 109 (1987).
- ²⁴M. Götz, K. Blüthner, W. Krech, A. Nowack, T. Wagner, G. Eska, K. Hecker, and H. Hegger, *J. Appl. Phys.* **78**, 5499 (1995).
- ²⁵Y. Nakamura, D. L. Klein, and J. S. Tsai, *Appl. Phys. Lett.* **68**, 275 (1996).
- ²⁶L. J. Geerligs, M. Peters, L. E. M. de Groot, A. Verbruggen, and J. E. Mooij, *Phys. Rev. Lett.* **63**, 326 (1989).
- ²⁷F. Deppe, S. Saito, H. Tanaka, and H. Takayanagi, *J. Appl. Phys.* **95**, 2607 (2004).