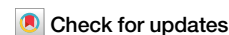


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Alternating-bias assisted annealing of amorphous oxide tunnel junctions



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Superconducting quantum bits (qubits) rely on ultra-thin, amorphous oxide tunneling barriers that can have significant inhomogeneities and defects as grown. This can result in relatively large uncertainties and deleterious effects in the circuits, limiting the scalability. Finding a robust solution to the junction reproducibility problem has been a long-standing goal in the field. Here, we demonstrate a transformational technique for controllably tuning the electrical properties of aluminum-oxide tunnel junctions. This is accomplished using a low-voltage, alternating-bias applied individually to the tunnel junctions, with which resistance tuning by more than 70% can be achieved. The data indicates an improvement of coherence and reduction of two-level system defects. Transmission electron microscopy shows that the treated junctions are predominantly amorphous, albeit with a more uniform distribution of alumina coordination across the barrier. This technique is expected to be useful for other devices based on ionic amorphous materials.

The preparation of amorphous thin film materials is surprisingly simple. While devices incorporating these materials are ubiquitous, their morphology is surprisingly complex and challenging to control. Among other similar materials, amorphous aluminum oxide (a-AlO_x) tunnel junctions stand out due to their importance in both cryogenic and room temperature devices. These applications range from those that utilize the Josephson effect¹, such as SQUID magnetometers², superconducting qubits^{3,4}, amplifiers^{5,6}, RSFQ logic⁷, to room temperature magnetic tunnel junctions⁸ and memory elements^{9–11}.

It has traditionally been assumed that the structural and transport properties of these amorphous materials are frozen-in once they are grown, and that high temperature annealing is required in order to modify them. Here, we demonstrate an alternating-voltage based technique, i.e. “Alternating-Bias Assisted Annealing” (ABAA), to re-order the atoms in these materials at relatively low temperatures in order to tune the electrical properties. Superconducting devices are used to demonstrate the efficacy of this technique. This is because, due to the cryogenic application of these oxide barriers for Josephson junctions, we can probe not only their resistance with high accuracy (using frequency), but also the prevalence of defects and loss using superconducting qubits. This is especially relevant to the field of quantum information since a-AlO_x is a key ingredient in transmon qubits¹², the invention of which has fueled much of the rapid development of superconducting applications research over the past decade.

The junction properties are crucial in the overall properties and performance of the qubit. Most importantly, the critical current, I_c of the barrier sets the Josephson energy of the device, $E_J = I_c \Phi_0 / 2\pi$, where Φ_0 is the flux quantum. E_J determines the parameters of the transmon such as the frequency and charge noise sensitivity. I_c can be predicted by the Ambegaokar-Baratoff formula, i.e. $I_c = \pi \Delta / 2eR_n$ ¹³, where, the variables R_n and Δ are the resistance of the junction (in its normal state) and the superconducting gap of the electrodes.

Unfortunately, while it is essential that I_c , and hence R_n , is well defined, a large spread of R_n in nominally identical junctions is typically observed, on the order of 1–10% at the wafer level¹⁴. This is due to both intrinsic properties and extrinsic effects. Intrinsic issues include the exponential dependence of the tunneling on the thickness¹⁵, grain structure, and interface roughness. Extrinsic effects arise from the effects of lithography uncertainties¹⁶, processing, electrostatic sensitivity, and circuit design.

Moreover, the amorphous nature of the oxide layer results in the inclusion of defects that can cause two-level systems (TLS's) near the qubit frequency that typically reduce coherence, or the defects can result in two-level fluctuators (TLF's) at very low frequencies that push the qubit frequency around^{17,18}.

The default method of making junctions, also used in this work, employs thermal oxidation of an aluminum electrode surface and shadow evaporation¹⁹. The accepted model for the oxidation process is described by

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the Cabrera-Mott model, illustrated in Fig. 1a, in which the aluminum/oxygen kinetics in these ionically bonded materials²⁰ is driven by an intrinsic electric field^{21,22}. This process is convenient and powerful because it is conformal and self-limiting in thickness. In the thin-barrier limit the formation of the oxide barrier is governed by drift-dominated atomic diffusion^{21,23} of the charged constituent elements, Al^{3+} and O^{2-} , in opposite directions, as illustrated in Fig. 1a. This diffusion is driven by the strong E -field ~ 1 GV/m generated by the adsorbed ionic ad-layer. In these, and other metal-insulator-metal oxides, the relevant parameter is referred to as the Mott voltage, $V_{\text{Mott}} \approx 0.5\text{--}1$ V for aluminum oxide²⁴.

There have been many efforts to improve the properties of the a- AlO_x material, focused primarily on understanding and optimizing the junction formation and post-processing, i.e., pressure, time²⁵, interface preparation, etches, simulations, etc., see ref. 26–28 and citations therein. These efforts have met with limited success, and it is typically assumed that the properties of the junctions, as formed, are relatively frozen-in and difficult to adjust. For example, annealing the entire wafer to high temperature after defining the junctions can significantly change the resistances, however, this approach has limited efficacy²⁹. This is due to a highly variable response of the various junctions, presumably due to nanoscopic morphology differences of nominally identical junctions at the domain and atomic level. Subsequently, methods of annealing individual junctions have been developed^{30–35}. These methods typically are implemented by targeting a slightly lower resistance in fabrication and then locally heating the individual junctions with high-power lasers or an electron beam to bring them into tolerance. However, these procedures involve the addition of complicated optical and imaging equipment to the test and measurement apparatus, and appear to be limited to adjustments of the resistances up to about 10–12% in Al/a- AlO_x /Al junctions, i.e. on the order of the junction spread. This limits the number of junction arrays that can be successfully adjusted, based on the initial distribution of the junctions.

On the other hand, it has been shown that oxidation can be enhanced by an external voltage during growth (by e-beam bombardment at the surface) or in capped structures, see ref. 36 and ref. 37, respectively. In addition, other physical material systems, e.g., magnetic³⁸ and piezoelectric³⁹, can have their properties improved by applying an alternating field polarity^{8,40}. While early work on the Al/Pb system, ref. 41, showed promising

results on charge motion in- and out-of the junction, trapping and other modifications of AlO_x using an alternating technique, no systematic approach to trimming the resistance and controllably modifying Al/ AlO_x /Al junction morphology has been investigated and implemented.

Results and discussion

In this work, we investigate the ABAA technique, illustrated in Figs. 1b and 2, to modify the resistance of thin amorphous tunnel junctions. The ABAA technique entails applying an alternating polarity voltage pulse train to the as-grown junctions during a global, low-temperature anneal. This is in sharp contrast to results obtained using only unipolar voltage pulses. For example, as shown in Fig. 2d, when unipolar pulse trains were applied to the junctions a relatively small change in the resistance was observed. Changes of about 2% and 6%, respectively, were registered for negative-only and positive-only voltages after about 200 pulses. However, when alternating-polarity-bias (AB) pulses were used, as illustrated in Fig. 2c, resistance changes of more than 70% were recorded as shown in Fig. 2d. In addition, we note two surprising features of the ABAA process that we address below: first, that a cyclic response was typically observed for the AB pulse trains, as illustrated in the Fig. 2d inset; and second, a systematic jump in the resistance curve typically occurs during the process.

Several features of Fig. 2d are instructive. For example, the asymmetry between the response to negative and positive pulse trains can be explained by considering the Cabrera-Mott mechanism for the oxidation. Since the initial oxidation involves only a negative bias, due to the charge transfer to the adsorbed oxygen, then relatively little change is expected when we re-apply that with the deposited top electrode. This can also create frustration, whereby cations and anions find themselves in metastable positions. When the voltage is reversed, anions and cations in the amorphous matrix can move more freely in the opposite direction, finding more stable positions, thereby allowing for a change in the resistance, as explained by the molecular dynamics simulations in ref. 26.

Moreover, when the bias is sequentially alternated, the two species can then begin to migrate and diffuse, creating a mixing effect that apparently accentuates this effect, leading to an anomalously high increase in resistance. This process is analogous to, for example, degaussing a magnetized sample, indicating that the junctions are slightly polarized even after adding the top electrode. This prediction is substantiated later in this work in the imaging section.

In addition, the cyclic, stepped response as the alternating process proceeds can be attributed to differential relaxation, not unlike in high-field pulsed magnetic samples⁴² with spin-glass properties.

This behaviour is consistent with the discontinuities observed in Fig. 2d, as well as in Fig. 3 after fewer and fewer pulses (i.e. shorter times) as the temperature is increased. The discontinuities could be explained by the hypothesis that the effective temperature of the sample is higher due to the ABAA, and the junction undergoes a phase transition from amorphous to glass, as described in ref. 43 for $T \sim 400^\circ$.

The temperature dependence of the ABAA from Fig. 3 also shows that the rate of resistance enhancement increases significantly as the temperature goes up. The increase is on the order of $2 \times$ every 10°C , consistent with that expected from an Arrhenius law. The ABAA effect was observed on all of the junctions in this study, with suppressed response observed from devices that had previously been heated above room temperature. Moreover, the size dependence (shown in Supplementary Note 1 and Fig. 1) has relatively small correction over nearly an order of magnitude in area, indicating that the mechanism is likely not due to the perimeter or other edge effects such as diffusion and/or surface migration. Rather, this points to local effects such as reordering and increased coordination of the atoms inside the barrier. This can lead to changes in the effective barrier height as atoms reach a more stable configuration²⁶. Furthermore, since the data was taken at constant voltage, we infer that it is not a current-driven diffusion effect. These data hold significant implications from the perspective of SQUID-based devices, such as sensors and tunable qubits, because these devices are typically made using two parallel junctions that may or may not be the same size.

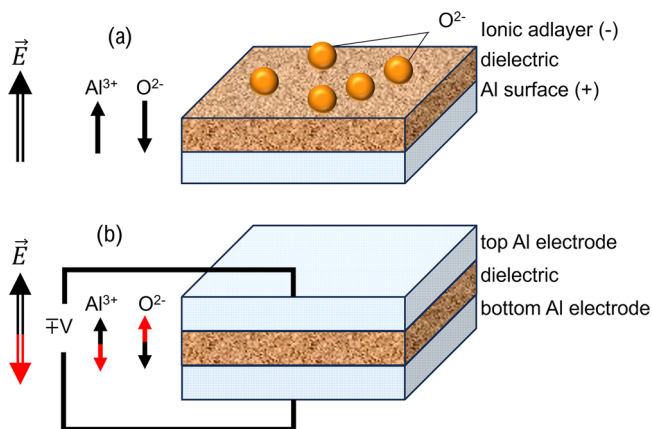


Fig. 1 | Models for thin-film oxidation of metals and ABAA. **a** Static Cabrera-Mott Model: Oxide growth on aluminum mechanism. The primary assumption is that adsorbed oxygen dissociates and ionizes. This creates a voltage V_{Mott} relative to the Al, causing a strong electric field in the thin oxide. Due to the predominantly ionic-bonded nature of these films (56%), aluminum cations and oxygen anions feel opposite forces due to this field. They exchange places, mediated by vacancy motion. **b** Dynamic Cabrera-Mott Effect (ABAA): The Al/ AlO_x /Al trilayer is effectively mixed by applying alternating voltages onto the tunnel barrier electrodes. The voltage is on the order of magnitude of V_{Mott} but less than the dielectric breakdown voltage, between the two plates. This applies alternating and opposing forces on the two species.

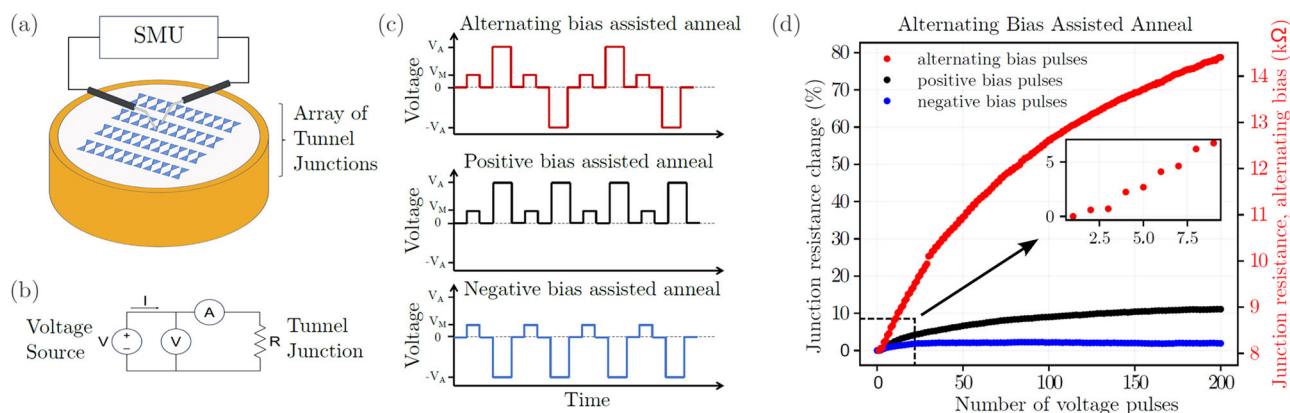


Fig. 2 | Schematic diagram of the bias schemes used to anneal tunnel junctions. **a** A block diagram of the apparatus. In all cases, the voltage source has the positive terminal connected to the top electrode and the negative terminal connected to the bottom electrode sitting on the substrate. The chuck can optionally be temperature controlled to assist with tuning. Typically 2-point probes are used to land on the individual qubit capacitor pads, since the junction resistance in this case is much higher than the probes. The junctions are formed on intrinsic-Si with ground rings around each qubit to avoid cross-talk. **b** Circuit diagram for the ABAA technique. **c** Bias pulses at V_A are applied for 1 second. In this paper $V_A = 0.9$ V. Between the bias pulses a low measurement pulse of $V_M = 10$ mV is used to measure the low bias

resistance of the junction. **d** Data from three different junctions to illustrate voltage-assisted annealing using alternating bias assisted anneal (ABAA) at 80°C vs. unipolar (positive and negative w.r.t. the top electrode) voltage pulses. The maximum voltage amplitude $V_A = \pm 0.9$ V with a 1-second pulse length. The area of the junction under test is $1.55\ \mu\text{m} \times 0.24\ \mu\text{m} = 0.37(\mu\text{m})^2$. The inset is a zoom-in on the start of the ABAA process and shows the cyclic change in junction resistance with a repeated sequence of small and large resistance changes when applying alternating bias bipolar pulses. The uncertainty in the resistance measurements is less than the size of the points.

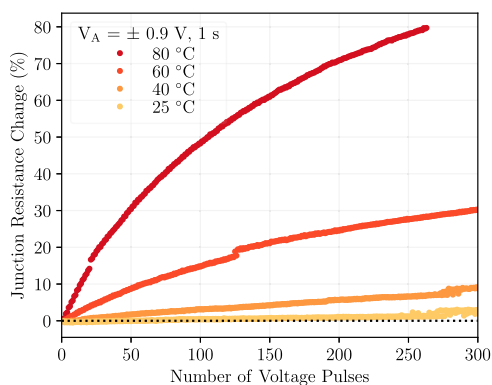


Fig. 3 | Junction resistance change as a function of the number of ABAA pulses. Data from four nominally identical junctions from the same wafer during the ABAA process at different substrate temperatures. In addition to the relatively smooth increase in junction resistance as the AB pulses are applied, discontinuities can often be observed as single events as in the process conducted at higher temperatures. In general, the annealing voltage amplitudes (for either polarity), pulse duration, and temperature are related. The resistance tends to saturate after many pulses during the process, however, under extended cycling the junctions can occasionally break down, consistent with nano-mechanical fatigue. After the process is complete, a slight overshoot of the resistance is observed, consistent and comparable to that reported for laser-annealing in ref. 33. The uncertainty in the resistance measurements is less than the size of the points.

We tested the effect of ABAA on the tunneling properties of the barrier by characterizing transmon qubits processed with and without ABAA. We measured a variety of properties associated with junctions used in both tunable and fixed-frequency qubits⁴⁴: whether I_C follows the behavior expected by the Ambegaokar-Baratoff relationship, Fig. 4; the presence of strongly coupled TLS's, fig. 5(a); and the impact of the barrier on qubit coherence, fig. 5(b).

The Ambegaokar-Baratoff parameters for this ensemble were first determined using the untreated devices. Then, the measured frequencies of the treated qubits were calculated for various resistance-trimming amounts. For the non-tunable qubits, this is straightforward, while for the tunable

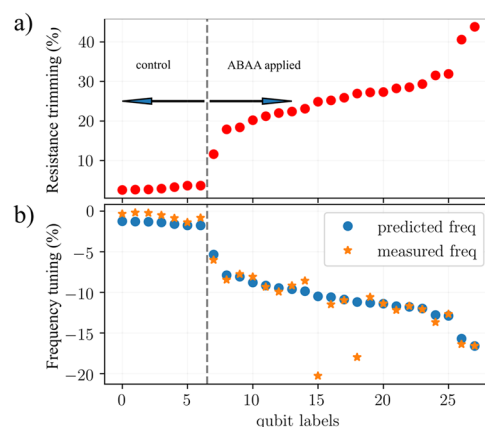


Fig. 4 | Translation of ABAA resistance trimming to frequency tuning.

a Measured junction resistance shift (red dots) resulting from the ABAA trimming of junctions. Control devices without the ABAA applied were included to confirm that the changes were due to the ABAA process and calibrate the Ambegaokar-Baratoff parameters, and ABAA was applied to the rest of the junctions. **b** The blue dots show the predicted frequency tuning from the resistance before and after the process for both the control ABAA treated junctions. The orange stars show the actual, measured frequency tuning of each qubit relative to the predicted values. Prediction error is typically 20 MHz, giving error bars that are about the size of the points.

qubits, it is necessary to find the maximum frequency, i.e. zero flux-bias, point using the qubit spectroscopy, shown in Fig. 5a.

Figure 4a shows the resistance change on several of the control qubits that were only processed at 80°C and the 21 ABAA-processed qubits. The ABAA processed chips had typical junction sizes, from ~ 0.05 to $\sim 0.35\ \mu\text{m}^2$. The junctions are ordered by the amount that their resistance changed during the processing. Critical currents from 20 to 30 nA for the treated junctions were extracted along with the charging and Josephson energies of the qubits (see Supplementary Note 2 and Fig. 2). It can be seen in Fig. 4b that the predicted and measured frequency tuning for ABAA-treated qubits match well, within $0.9 \pm 2.4\%$, demonstrating that the trimmed resistances, as-measured after ABAA processing, can be used in the

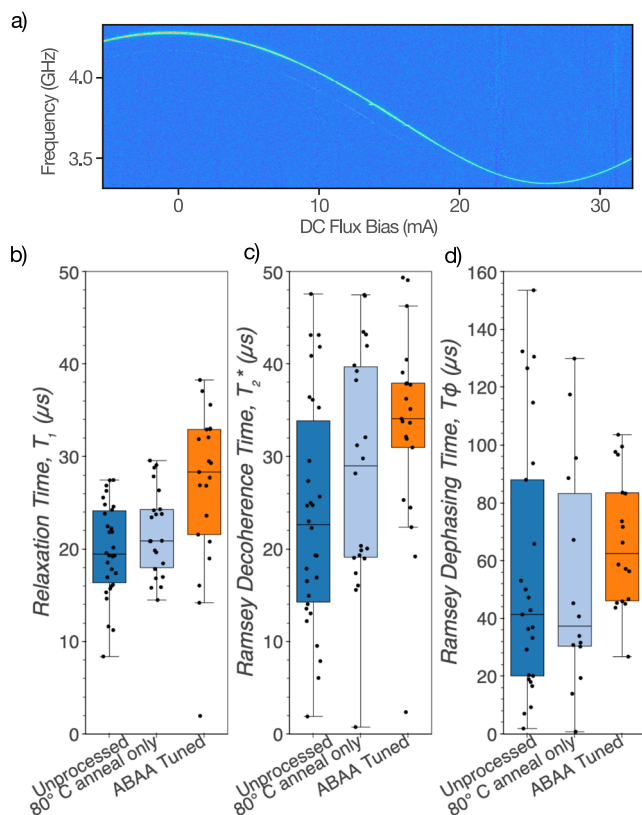


Fig. 5 | Spectroscopic and coherence properties of the qubits. **a** A representative spectrum of a tunable qubit that has been treated with ABAA. The two small features at approximately 3.8 GHz and 3.7 GHz correspond to neighboring qubits. **b–d** Qubit coherence metrics T_1 , T_2^* , and T_ϕ , respectively. The study included two types of controls, i.e. multiple unprocessed qubits and qubits heated to 80 °C only. The ABAA tuned qubits were processed at $V_A = \pm 0.8$ V, 1 s and $T = 80$ °C. The filled boxes error boxes span the 1st and 3rd quartile of the data uncertainty, while the whiskers mark the furthest datum within the $1.5 \times$ the interquartile range past the 1st and 3rd quartiles.

Ambegaokar-Baratoff formula. We find that this is consistent between junctions with ABAA applied vs. unprocessed with values of (1.32 ± 0.084) pH/ Ω and (1.27 ± 0.109) pH/ Ω respectively.

The success rate of the ABAA-process for the initial tests on a process-test wafer was 79%, as discussed in Supplementary Note 3.1. In a subsequent extended study to optimize the ABAA process, a yield of $97.4 \pm 0.4\%$ was observed, see Supplementary Note 3.2.

The qubit spectroscopy also probes for the existence of avoided level crossings in this frequency range. Such crossings are signatures of strongly coupled TLS defects within the tunnel barrier^{45,46}. For reference, measurements on untreated qubits show about 0.7 TLS/GHz, and on those that were heated to 80 °C without ABAA have 0.6 TLS/GHz. Compared to these reference measurements, for the nine tunable devices in the ABAA-treated ensemble of this study with 5.4 GHz of tunable range, we saw no evidence of avoided level crossings larger than our ~ 500 kHz resolution. Figure 5a shows typical spectroscopy from an example qubit. Several other examples of TLS spectroscopy, both treated and not, from the ensemble are discussed in Supplementary Note 4, Figures 3–6. We note here that smaller splittings, typically detected with swap spectroscopy⁴⁶ and likely due to surface defects, would not be affected by the ABAA. While this small data set is not conclusive evidence that such defects are suppressed by the ABAA process, it does point to the possibility that ABAA may improve qubit coherence and frequency stability. For example, the low-frequency stability of one of the devices was monitored over many hours (see Supplementary Note 5 and Figure 7), and there was no evidence of coupling to TLFs¹⁸. In order to investigate this issue further, we studied the loss and time domain coherence in the ABAA qubit ensemble.

The coherence metrics are shown in Fig. 5b–d for all qubits measured in the initial study, and corresponding loss is discussed and shown in the Supplementary Note 6 and Table 1. Here, each data point corresponds to the median of at least 60 measurements on each qubit in the ensemble of ≈ 70 qubits. (see discussion in Supplementary Note 3.1) We observe that the ABAA processed devices have a higher median T_1 than both the unprocessed devices and the devices that were only heated to 80 °C, although the T_2^* and T_ϕ values between sample sets are not distinguishable within the spread of the values.

When analyzing processes that may reduce loss in qubits, it is useful to compare these metrics in the form of a qubit decay rate normalized to frequency $[(2\pi f_{01} T_1)^{-1}]$; the ABAA tuned sample set has median loss tangent of 1.4×10^{-6} , while both the 80 °C annealed and unprocessed sets have median loss tangents of 1.7×10^{-6} . We found that the coherence was typically improved after the ABAA process, indicating that the resistance trimming may have reduced the median loss tangent in the qubits by about 0.3×10^{-6} . These qubits are similar to devices in ref. 47 and “Design B” from ref. 48, where the junction has relatively low participation. Hence, the modest increase in T_1 reflects the electrode surfaces/materials-centric design of these qubits.

Finally, transmission electron microscopy (TEM) imaging and scanning-TEM (STEM) with electron energy-loss near-edge structure (ELNES) analysis and high-angle-annual dark-field (HAADF) imaging, respectively were employed. These allowed us to investigate morphological changes in the junctions due to the ABAA process. Details for this study are provided in Supplementary Note 7. These studies indicate that the increase in junction resistivity may be attributed to a more uniform barrier structure with reduced point defect density. A low-mag cross-sectional TEM/STEM image and corresponding energy dispersive spectroscopy (EDS) elemental distribution map are shown in Fig. 6a–c. This demonstrates that the a- AlO_x barrier is still amorphous in the ABAA-treated sample (Fig. 6d), and the HAADF-STEM image shows a barrier thickness of approximately 2 nm (Fig. 6e).

While the ABAA technique does not appear to change the amorphous nature of the barrier from visual inspection, the chemical analysis tells a different story. We see a more homogeneous chemistry in the ABAA-treated a- AlO_x than in untreated junctions. Our previous research showed that the distance between two main peaks labeled as A and B in Fig. 6f–g, close to approximately 77.5 eV and 80 eV of the AlL2,3 energy-loss near edge structure (ELNES) respectively, can be used to analyze the coordination number of Al^{49–54}. Specifically, the Al-L3 edges (peak A) tends to shift to higher energy as Al coordination numbers increase. For untreated junctions, a spatial variation in coordination number across the a- AlO_x barrier from the untreated junction (Fig. 6g)⁴⁹ is demonstrated and considered as a potential decoherence source. In contrast, the ABAA-treated junction shows a uniform distribution of Al coordination number through the barrier, as the peak positions of A and B remain unchanged across the a- AlO_x layer, as shown in Fig. 6f. This may originate from the migration/recombination of charged point defects induced by the alternating bias field⁵⁵ giving rise to a more chemically homogeneous barrier layer. This effect may increase the effective barrier height of the tunnel junction, leading to the observed increase in resistance.

Conclusion

We find that an alternating bias is effective in assisting the thermal annealing of ionically-bonded amorphous materials. The effect is attributed to depolarization of the as-fabricated amorphous matrix as cations and anions are driven in opposite directions, allowing them to find more stable positions, similar to adding restarts in simulated annealing solvers to avoid local minima⁵⁶. This allows for significant modifications of the junction resistance, in excess of +70%, with apparent reductions in loss and defects. Junctions that were tested, however, we do not These modifications can be made easily with a standard probe station at the wafer, die, or device level. In practice, we find the exponential increase is stable and tends to saturate at a high number of pulses. The behavior of the resistance during the ABAA

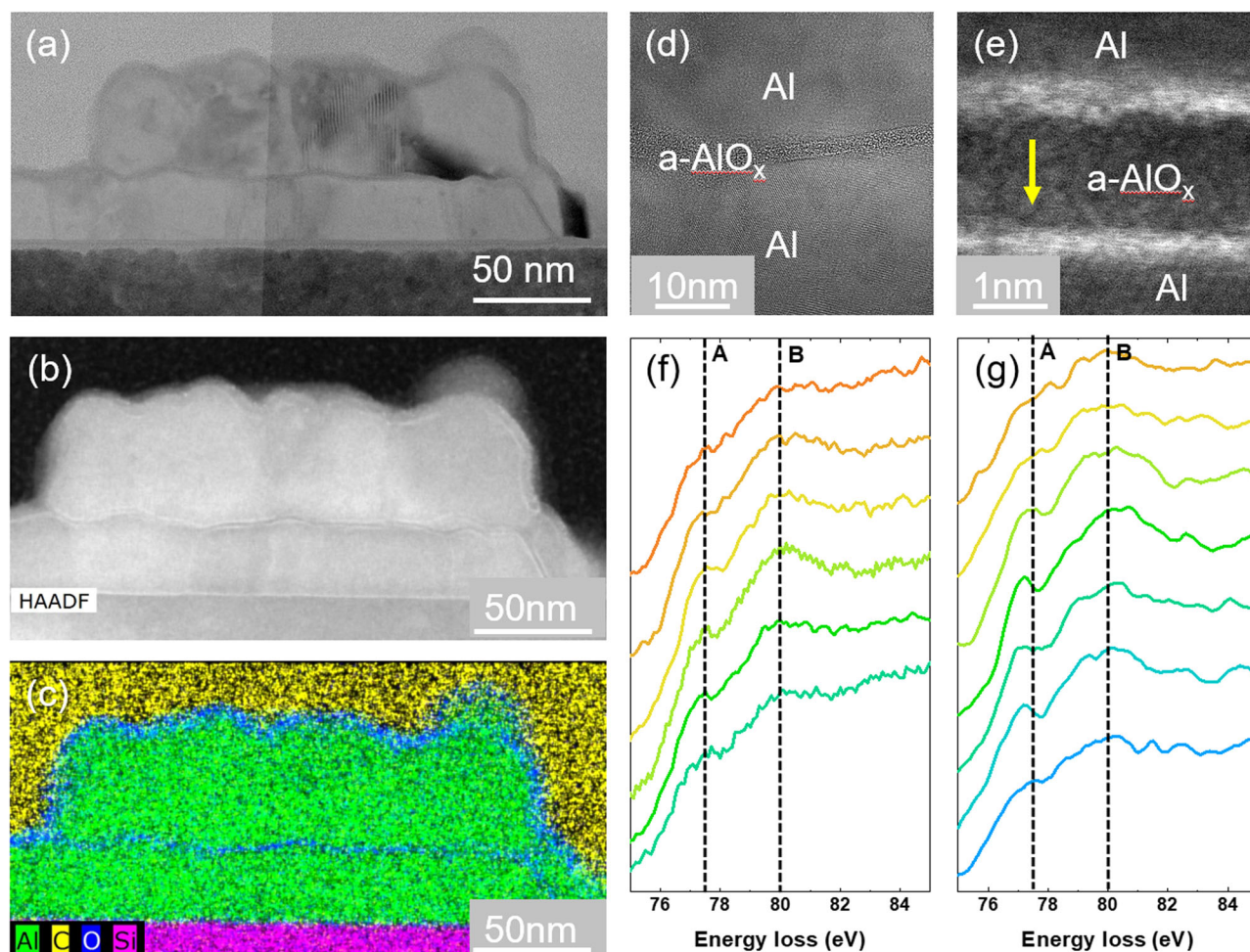


Fig. 6 | Electron microscopy analysis on ABAA-treated junction. **a** Low magnification cross-sectional TEM image of a junction. **b** HAADF-STEM image of the same junction. **c** Corresponding STEM-EDS elemental distribution map of the junction. **d** High-resolution TEM image of the Al/a-AlO_x/Al interfaces. **e** HAADF-

STEM image of Al/a-AlO_x/Al interfaces. **f** ELNES of Al-L2,3 edge of the ABAA-treated sample taken from the yellow arrow indicated region in **e**. **g** ELNES of Al-L2,3 edge of the untreated a-AlO_x barrier from reference⁴⁹.

process showed cyclic small and large changes in resistance as the positive- and negative-bias voltages were applied and the junction resistance increased, and typically a single discontinuity in resistance was observed if a sufficient number of pulses were applied.

These results, along with the temperature-dependent studies and imaging, point to an assisted annealing mechanism. This appears to have improved local ordering and/or the a-AlO_x being driven into a lower-energy glassy state^{43,57,58} in the potential energy landscape, as indicated by the observed reduction in loss and TLS. In order to confirm these hypotheses, more studies such as high-resolution imaging and molecular dynamics that include density-functional theory are required. Regardless of the mechanism, we expect that a controlled alternating bias-assisted junction annealing approach will pave the way for building large-scale superconducting quantum processors by improving qubit frequency targeting, coherence, and stability. Other applications, where targeting tunnel junction resistance is important, may benefit by allowing for more homogeneous junction chains and ensembles for devices ranging from amplifiers to voltage standards, sensors, and data storage.

Methods

The ABAA treatment was implemented on as-grown junctions using a conventional probe station with a low-temperature heating stage, as illustrated in Fig. 2(a). Using this technique, it can be expected that the Al³⁺

cations and O²⁻ anions in the barrier will be driven in opposite directions at each pulse. By alternating the bias voltage the ions can be driven out of local, metastable sites into a lower energy configuration. In general, the guidelines used for the voltage profiles were that it should be on the order of V_{Mott} and less than the breakdown voltage, $V_B \sim 1.5 \text{ V}$ ^{11,24}, as discussed and illustrated in Supplementary Note 8 and Fig. 9, respectively. When considering the time duration of the pulse, we note that by staying well below V_B the junction properties were observed to evolve relatively slowly, on the order of hundreds of seconds, similar to the known oxidation rates of these surfaces. Finally, a relatively low temperature is desired to not expose junctions to a higher temperature than the highest temperature they are exposed to during the fabrication process.

The equipment and pulse sequences used for the tuning consist of a standard source-measure unit (SMU) and a probe station with a heated platen, illustrated in Fig. 2a, b. The SMU was programmed to provide a train of 1-second-long anneal-assist pulses after the wafer was brought to the target temperature. The pulse amplitudes were set in the range of 0.8–1.1 V. The resistance of the junctions was measured after each pulse with an additional pulse at low voltage.

Data availability

The datasets generated during and/or analysed during the current study are available from the corresponding authors on reasonable request.

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Author contributions

D.P.P. designed the experiment and wrote the manuscript; M.F. and X.W. made ABAA measurements; C.J.K. identified and provided samples and qubit measurement results; J.A.H. conducted analysis and frequency predictions; E.S. provided theoretical support; E.L. and A.G. conducted literature searches and provided critical input for the manuscript; L.Z. and J.O. conducted and interpreted microscopy; G.M.S. conducted and interpreted qubit measurements; A.B., K.Y., and M.J.K. supervised the work; J.Y.M. motivated and supervised the work. All authors discussed the results.

Competing interests

D. P. Pappas, M. Field, C. Kopas, J. A. Howard, X. Wang, E. Lachman, K. Yadavalli, Allyson Gold, E. A. Sete, A. Bestwick and J.Y. Mutus are, have been, or may in the future be participants in incentive stock plans at Rigetti Computing Inc. D. P. Pappas, M. Field, X. Wang, E. A. Sete, and J.Y. Mutus are inventors on a pending patent applications related to this work (No. 63/612,176). The remaining authors declare that they have no competing interests.

Additional information

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