

Analysis of Josephson junction barrier variation: A combined electron microscopy, breakdown, and Monte Carlo approach

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Josephson junctions manufactured to tight tolerances are necessary components for superconducting quantum computing. Developing precise manufacturing techniques for Josephson junctions requires an understanding of their make-up and robust feedback metrics against which to optimize. Here, we consider complementary techniques and assess what conclusions they allow us to draw about the barriers in junctions. Monte Carlo simulations of barriers show that standard deviations of 15–20% of the total barrier thickness are compatible with our experimental data. Electrical breakdown allows us to probe the weakest points in barriers. Narrowing the distribution of this breakdown provides a promising feedback mechanism for barrier optimisation. Grouping junctions by breakdown voltage allows us to identify sub-ensembles of junctions with different median resistance. Transmission electron microscopy can be used to find average barrier thickness, although we highlight challenges in forming robust conclusions on the distribution of thicknesses in a barrier from these experiments.

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I. INTRODUCTION

Josephson junctions (JJs) are ubiquitous to superconducting quantum computers [1] and other superconducting circuits such as parametric amplifiers [2] and therefore have significant technological importance. The most widely used type of JJ in superconducting technologies is based on an insulating layer of AlO_x , which separates two superconducting electrodes. The AlO_x layer is formed by oxidizing the surface of a layer of metallic aluminum before covering the AlO_x layer with a second superconducting electrode, also aluminum. This creates a barrier that is (i) amorphous, (ii) nonstoichiometric, and (iii) has a thickness that varies across the junction [3–6]. Developing manufacturing techniques for JJs with fine tolerances requires a detailed understanding of the junction, as well as robust feedback metrics against which manufacturing techniques can be optimized.

Typically, when designing superconducting devices reliant on JJs, a specific critical current value I_C is targeted. Ensemble measurements of I_C are typically used to characterize Nb/ AlO_x /Nb JJ properties [7–9]. This property can be harder to directly measure across statistically significant ensembles in Al/ AlO_x /Al JJs because of their critical temperature being substantially below liquid helium temperatures. A proxy for this value in junctions with clean insulating barriers is the room-temperature resistance [10,11] and resistance

tolerances are typically used to characterize manufacturing processes. These processes are in turn optimized considering feedback from resistance measurements and techniques such as scanning electron microscopy [12,13]. Consequently, many problems with repeatable JJ manufacture have been solved and control of the area of JJs is now much improved [10,14]. As a result, the current state of the art in as-fabricated JJ resistance spread is $\sim 2\text{--}3\%$ [15–17]. Once lateral dimensions of junctions are perfectly controlled, the remaining variation between junctions must be attributed to differences between their barriers although it is not clear what level of resistance variation this causes in typical manufacturing processes.

Variability in junction barriers has more complex consequences on the junction physics than being a poorly controlled variable that rescales the critical current affecting manufacturing tolerance. Nonideal JJs can be characterized by their expanded current-phase relationship ($C\phi R$). In these realistic junctions, the first Josephson equation, $I = I_C \sin(\phi)$ where ϕ is the superconducting phase drop across the JJ [18], becomes more complicated with current given by the Fourier series $I = \sum_n I_{C,n} \sin(n\phi)$ [19]. Imperfections in the barrier cause variations in $I_{C,n}$, changing the critical current, and adding higher-order terms to the $C\phi R$ [20]. These higher-order terms can then affect the properties of devices based on these JJs [21].

Variation in dielectric thin films, like the barrier, is a well-studied problem with relevance to devices including field-effect transistors [22], metal-insulator-semiconductor or metal-insulator-metal structures [23,24], and magnetic tunnel junctions [25]. The barriers in JJs are typically $\sim 1\text{--}2\text{ nm}$ [26], so even a single O^{2-} ion represents a substantial fraction of the average barrier thickness with ionic radius of $\sim 0.14\text{ nm}$ [27]. Generating a full 3D map of barriers at this resolution is not yet possible although various techniques have been used to improve understanding of barriers. (Scanning) transmission electron microscopy [(S)TEM] and associated techniques

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give chemical and structural information at subnanometre resolution [4,28]. Atom probe tomography results in 3D reconstructions of the barrier although, to date, results on AlO_x barriers do not show the requisite resolution [28,29]. Scanning tunneling microscopy (STM) maps the as-grown oxide prior to being capped by a final metal layer [25], and oxygen migration occurs after this final capping [30]. Simulations of the growth process by molecular dynamics give insight into atomic structures in typical barriers [30]. Studies of breakdown voltages of barriers are used in device communities, such as field-effect transistors, to probe device-to-device variation in barrier weak points [31], although they have not seen substantial use in JJs.

In this work, we consider electrical measurements and STEM imaging of barriers, discussing what conclusions can be robustly drawn from these types of characterization. We extend the typical range of electrical measurements to sample nonlinear parts of the current-voltage (IV) curves and junction breakdown. We complement this by Monte Carlo simulations of IVs and breakdown resulting from nonuniform barrier thickness. We are able to recreate the extended measurements with these Monte Carlo simulations considering only statistically distributed thicknesses within single junctions and find that skewed thickness distributions fit our experiments best. We then consider how thickness information is derived from STEM imaging, a technique that has previously been used to measure thickness distributions in barriers. We consider how choices in image processing and nonidealities in the barrier electrodes may affect conclusions drawn from this type of analysis.

We study tunnel barriers made by shadow angle evaporation [32] of aluminum forming $\text{Al}/\text{AlO}_x/\text{Al}$ tunnel barriers, which, when cooled to cryogenic temperatures, form Josephson junctions. Using the same processes as in [16], junctions are defined in a Dolan bridge geometry [32] via electron beam lithography. A base layer of aluminum is evaporated onto a sapphire substrate and is then oxidized in a static oxidation process before being capped with a top layer of aluminum and lifted off. All junctions in this work are fabricated on a single sapphire wafer in one fabrication run.

II. ELECTRICAL MEASUREMENTS

Figure 1 shows results from fitting a set of 597 IV curves. These are collected from junctions manufactured with the same target dimensions. An atomic force microscope (AFM) measurement of one of these junctions is shown in Fig. 1(a), from which we find the junction dimensions to be $\sim 240 \times 240 \text{ nm} = 5.76 \times 10^4 \text{ nm}^2$. We apply a voltage across each junction, which is swept from 0 V to a value up to 1.6 V as shown in Fig. 1(b). At low voltages, the IVs of the JJs are linear and we fit this dependence at voltages below 0.02 V to extract the junction resistance. At voltages below 1.6 V, each junction fails at its breakdown voltage and becomes an ohmic channel with much lower resistance (typically hundreds of Ohms).

We also fit the IVs for all voltages below the breakdown voltage using the Simmons model, which gives the current-voltage properties for a thin rectangular tunnel junction as a function of the potential barrier height and its

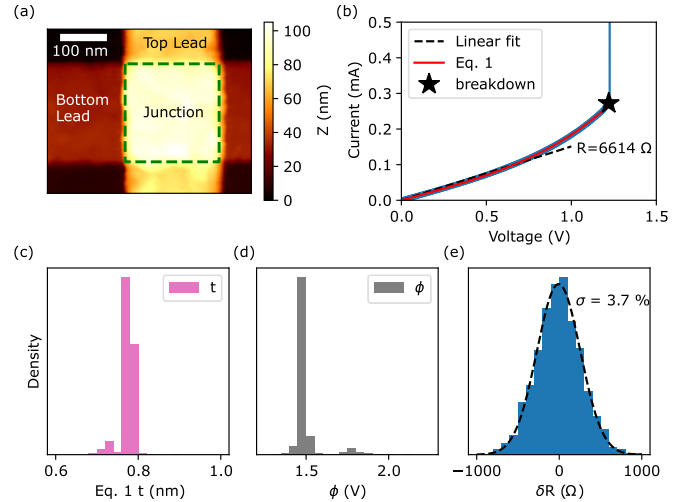


FIG. 1. (a) An AFM measurement of a JJ from the ensemble of JJs. (b) An IV of a typical junction showing the extraction of the breakdown voltage, resistance from a linear fit to the low-voltage regime and a fit to the full Simmons model. (c), (d) Results from analyzing an ensemble of 598 junctions fabricated on a 3" wafer. (c) A histogram showing the outputs from fitting Eq. (1) to measured IVs with thickness, barrier height, and nominal area shown on the same x axis. (d) A histogram of the resistance deviation from the median resistance of the junction ensemble with a median resistance value of 7122Ω .

thickness [33],

$$I = \frac{Ae^2}{2\pi\hbar t^2} \left[\left(\phi - \frac{V}{2} \right) \exp \left(-K \sqrt{\phi - \frac{V}{2}} \right) - \left(\phi + \frac{V}{2} \right) \exp \left(-K \sqrt{\phi + \frac{V}{2}} \right) \right] \quad (1)$$

where A is the junction area [constrained to the value found by AFM in Fig. 1(a)], t the junction thickness, ϕ the barrier height, and $K = 4\pi t \sqrt{2m_e e}/\hbar$. Fitting this model gives a single-valued thickness, which, as discussed in the introduction and shown in the literature, for example, by STM [25], is a simplification of the true barrier in these junctions. The simple analytical form makes it computationally inexpensive to compute, which is important when used in Monte Carlo simulations shown later.

In Figs. 1(c) and 1(d), we show the results of fitting Eq. (1) presenting t and ϕ , respectively. We find median $\pm$ standard deviation values of the fit parameters given by $t = 0.78 \text{ nm} \pm 3\%$, $\phi = 1.48 \text{ eV} \pm 6\%$. In Fig. 1(e), we show a histogram of the JJ resistances extracted from the linear fit, centered at 7122Ω with a normally distributed resistance and a standard deviation of 3.7%. We use these experimental results to define targets for Monte Carlo simulations of IV curves.

We perform Monte Carlo simulations where we allow thickness to vary across a junction trying to understand if we can recreate experimental measurements considering only thickness variations within junctions. To build these simulations, we define a junction with an area corresponding to the overlap between the two electrodes in a JJ (shown in Fig. 1(a) and within the Supplemental Material [34]). This

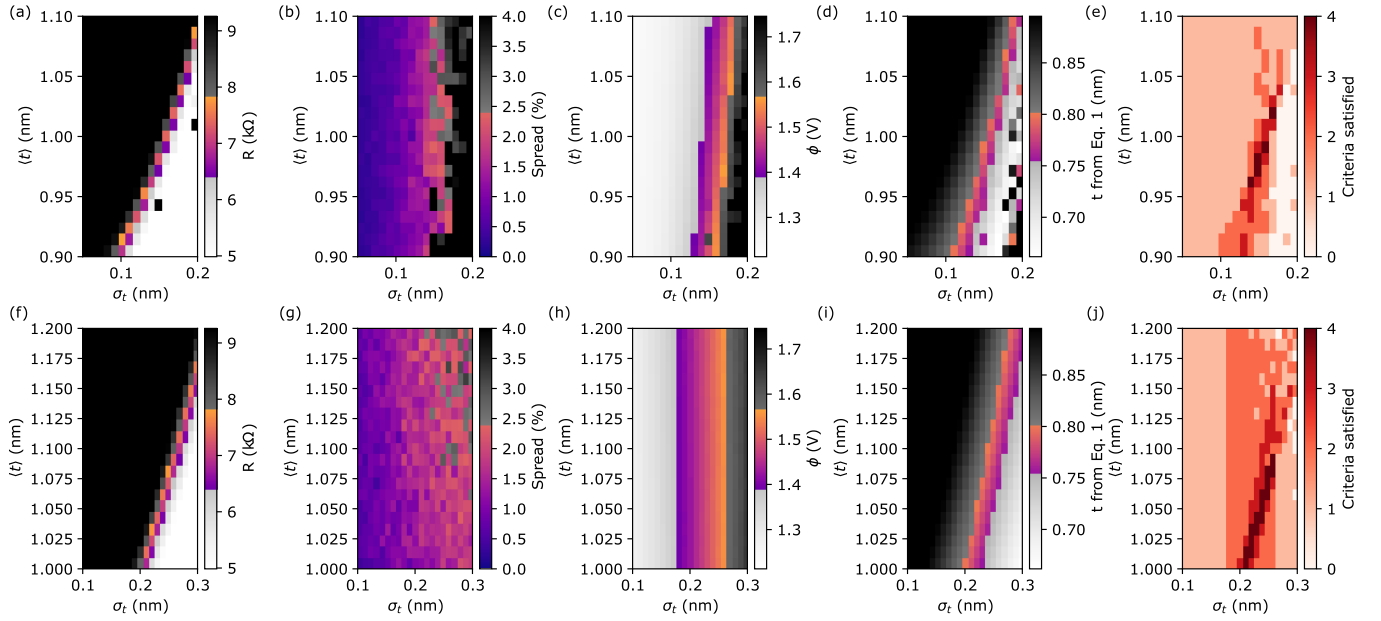


FIG. 2. Results from Monte Carlo simulations of barriers with normally (a)–(e) and lognormally (f)–(j) distributed thicknesses. Heatmaps of (a), (f) resistance, (b), (g) resistance spread, (c), (h) barrier height (ϕ), and (d), (i) fitted thickness. Resistance and its spread are found by fitting a straight line to the low-voltage region of the simulated IV and other parameters are found from fitting Eq. (1) to the full voltage range of the simulated IV. Any regions within the experimental variation (or 10% instead for resistance) are shown using a purple to orange heatmap, whereas values outside this range are shown in grayscale (e), (j). For each pixel, we sum the number of experimental criteria matched by the simulation. Dark hues indicate regions where the experiment and simulation coincide, indicating that there is a parameter regime for each distribution where this matching is achieved.

area is divided into equally sized pixels, which act as parallel conduction channels. The thickness of the barrier at each pixel is drawn from a statistical distribution and then Eq. (1) is computed as the IV for that pixel. Combining the IVs of each pixel as parallel conduction paths simulates the IV of the whole junction. We consider both normally distributed and lognormally distributed thicknesses, where a lognormal distribution is an example of a skewed thickness distribution. We characterize each simulated IV curve by refitting it to Eq. (1), which allows us to compare simulated IVs to experimental IVs. In the fitting routine, we constrain the area of the junction to be the same value as that found by AFM. Fitting the linear regime of the IV gives the resistance of the junction. Repeating this protocol for multiple junctions (20 in this work) gives the junction-to-junction variation for a given thickness distribution. The pixel size is an important parameter in these simulations with smaller pixels giving less junction-to-junction variation. Pixels less than ~ 0.2 nm become unphysical as they drop below ionic radii of the barrier [27], giving a lower bound on pixel size. We perform these simulations with a pixel size of 1 nm and a barrier height of $\phi = 1.22$ eV. The pixel size is chosen to correspond approximately to the thickness of barriers assuming that the barrier thickness does not change on a length scale much less than the thickness and also as a compromise for computational speed. The value of barrier height is important for inferring accurate thicknesses from any subsequent modeling. Literature values vary with changes in barrier height being ascribed to the termination of the oxide barrier (Al/O) and the crystal structure of the underlying metal [35,36]. Experimental studies also using the Simmons model identify barrier heights ranging from

~ 0.9 eV to ~ 1 eV [37]. Density functional theory (DFT) simulations of amorphous Al/AIO_x/Al barriers show dependence on roughness of the underlying metal and find effective barrier heights ranging from 0.2 eV to 1 eV [38].

We show the results of these Monte Carlo simulations in Fig. 2. We compare these to experimental results with different criteria for matching experimental results. Resistance is considered a match if the simulated value is within 10% of the experimental values. Resistance spread is considered a match if the spread is $\leq 2.4\%$, as presented in the Supplemental Information of Ref. [16] for junctions made with this recipe. Simulated values of t and ϕ are considered to match if they are within one standard deviation of the median value found by fitting ensembles of experimental IV curves as shown in Fig. 1. The top and bottom rows show results for normally distributed and lognormally distributed barrier thicknesses, respectively. In each heatmap, we color any cells compatible with the experimental data with a purple-orange heatmap, and show cells incompatible with experimental results in grayscale. For both distributions, there is a region where the experimental results shown in Fig. 1 are matched well by simulations indicated by the darkest hues in Figs. 2(e) and 2(j). Using a multivalued thickness within a junction, we are able to match experimental IVs whilst maintaining the known lateral dimensions of the junctions.

These simulations also show that inferring the barrier height by extracting the value from fits to the Simmons model is not straightforward. In Fig. 2, we use a single value of barrier height $\phi = 1.22$ eV. However, the value of ϕ extracted from refitting these simulations shown in Figs. 2(c) and 2(h) deviates from this value, depending on the underlying

thickness distribution. For instance, moving along the x axis for a fixed value of $\langle t \rangle$ in both Figs. 2(c) and 2(h), the barriers with greater thickness variation appear to have larger barrier heights. This may explain observations such as those seen in Ref. [37], where they conclude that oxidation time has changed the barrier height. Instead of changing barrier height, it may be that the oxidation time is changing thickness distributions (either average or variation), which appear as potential barrier heights when interpreted via fitting to the Simmons model. Given the variation in literature values and imperfect inference from electrical measurements, direct measurements of the barrier height would be advantageous, for instance by x-ray photoelectron spectroscopy techniques such as in Ref. [39].

To understand the effect of changing barrier height, we perform these simulations for $\phi = 1.5$ eV, 1 eV, and 0.8 eV, and show these in the Supplemental Material [34] and compare the results of fitting the Simmons model to data. With large barrier heights (similar to the median values found experimentally), we find a parameter regime where simulations match experiment. This parameter regime has very uniform barriers (barriers that vary less than an ionic radius). This is contrary to observations such as those in Refs. [40,41] where thickness variation can be seen at grain boundaries. For lower barrier heights, we do not find any regions where all parameters match the experiments. The parameters that match the most experimentally imposed conditions imply a thicker average barrier and also a larger thickness standard deviation. When using the barrier height of 0.8 eV, we find average thicknesses that match our best inference from STEM imaging discussed below. This places a lower bound on barrier heights compatible with our results. The use of a rectangular potential barrier will give a systematic offset, implying a thinner barrier relative to what would be inferred using more realistic rounded potential barriers. A detailed understanding of potential barrier shapes or atomistic computations of tunneling currents would improve this model and likely result in better quantitative agreement between simulated potential barrier thicknesses and the thicknesses of AlO_x barriers. We could also extend this model to draw ϕ from a distribution of values indicating compositional fluctuations at the expense of extra computational time. An understanding of realistic levels of potential barrier fluctuations would be necessary for this to give deeper insight into our results. The use of a single-valued potential-barrier height here is equivalent to an assumption that fluctuations in thickness contribute more to IVs than fluctuations in barrier height.

We can complement the information shown from the fitted IV curves with measurements of the breakdown voltage, the point indicated by a star in Fig. 1(b). We present a histogram showing the analysis of junction breakdown voltages of the same group of junctions in Fig. 3(a). We phenomenologically fit a double-Gaussian to the breakdown voltage, showing a bimodal distribution with a midpoint of 1.3 V. Breakdown occurs at the weakest point in the barrier at a voltage given by $V_{BD} = tE_{ds}$, where t is the barrier thickness and E_{ds} its dielectric strength. Assuming a uniform dielectric strength across the barrier, the weakest point is determined by the thinnest point in the junction. Dielectric strength could also fluctuate across the barrier, for instance decreasing at defects in the

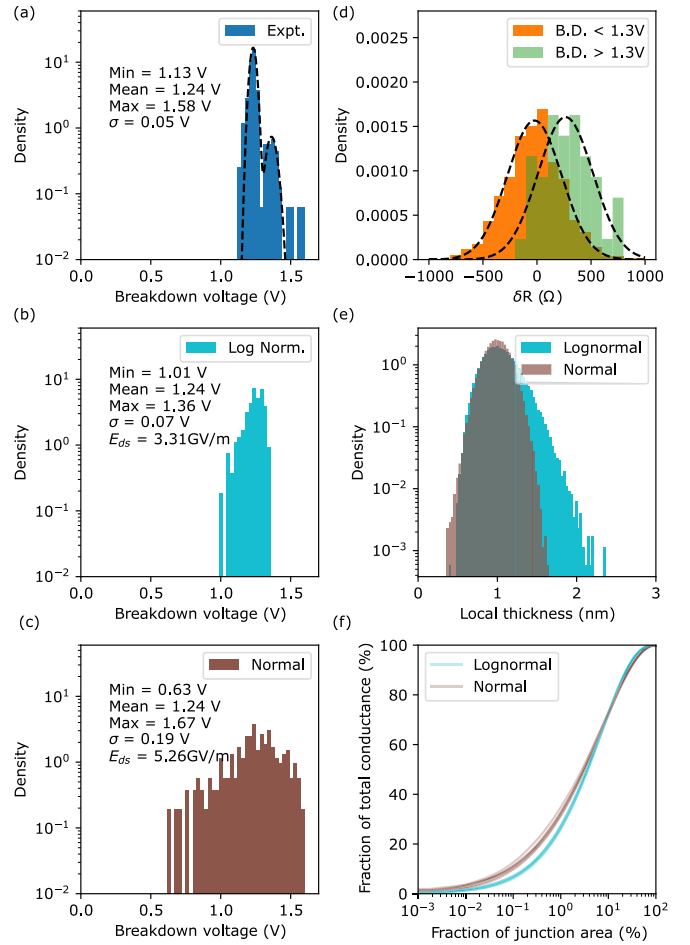


FIG. 3. Comparison of (a) experimentally measured breakdown voltages with simulated breakdown voltages for (b) lognormally and (c) normally distributed barriers created with distributions in regions where all experimental criteria are matched in Fig. 2. (a) Shows a double-Gaussian fit indicating a bimodal breakdown distribution. Simulated distributions are found by taking the thinnest points in randomly generated barriers with a mesh size of 0.2 nm and rescaling these by a dielectric strength (E_{ds}) to keep the mean breakdown voltage equal to the experimental values. (d) The resistance of junctions distinguishing junctions with breakdown above/below the midpoint of the bimodal distribution 1.3 V, shown by the black dashed line in (a). (e) A histogram of the full thickness distribution of example junctions generated with the parameters from (b) and (c). (f) Cumulative conductance of junctions from 10 different junctions generated with parameters from (e) as a fraction of the total junction area.

barrier, where what constitutes a defect in an amorphous oxide is a complicated issue in itself [42]. If all junctions have the same thickness at their thinnest point, then the spread in Fig. 3(a) would be a direct measure of the spread in dielectric strength, allowing us to place a bound on the standard deviation of this material property in these barriers, $\sigma_{E_{ds}} < 4\%$. We consider the case where dielectric strength is constant across all barriers and within each barrier, i.e., omitting any contributions from local structural changes. In this limit, the breakdown voltage is determined by the thinnest point of each barrier, and the histogram in Fig. 3(a) is a rescaled measurement of these thinnest points from 597 junctions.

Extending the Monte Carlo simulations shown in Fig. 2, we consider whether the same model of varying barrier thickness agrees with our experimental measurements of breakdown voltage. Using thickness distributions identified in Fig. 2 as giving the matches between simulated and experimental IVs, we compute a distribution of breakdown voltages. For normal (lognormal) distributions, we use a thickness standard deviation and average of 0.21 and 1.025 nm (0.155 and 1 nm), respectively.

We generate thicknesses from the distributions within the region of experimental matching in Fig. 2 for a 240×240 nm junction with a 0.2 nm pixel size [43]. We record the minimum thickness of the barrier in the junction and then repeat this to build a distribution of the thinnest points in the barrier. In order to translate the distribution of the thinnest points to breakdown voltages, we rescale the distribution of thin points by a constant dielectric strength of 3.31 GV/m and 5.26 GV/m for lognormal and normal barrier distributions, respectively. Previous measurements of E_{ds} in amorphous alumina vary from 0.4 to 1.0 GV/m, similar in order of magnitude [44]. We compute the dielectric strength by enforcing the mean of the rescaled thicknesses to be equal to the mean of the measured breakdown voltages. Simulated breakdown distributions for barriers with lognormally and normally distributed thickness are shown in Figs. 3(b) and 3(c), respectively. Neither statistical distribution recreates the bimodal distribution seen experimentally. However, the rescaled values from lognormally distributed barriers [Fig. 3(b)] show reasonably good agreement with the measured breakdown voltages, with a similar standard deviation and min/max ratio. In contrast, the rescaled values taken from barriers generated with a normally distributed thickness [Fig. 3(c)] do not agree with the experimental measurements. We show that multiple experimental measurements can be largely recreated considering only a skewed thickness distribution in our barriers. The parameters used to simulate breakdown are drawn from simulations of IV curves that have no awareness of breakdown physics, and only one value is used to rescale the distribution. We note that we do not recreate a bimodal distribution, indicating that, while better than a normal distribution, the lognormal thickness distribution is not a perfect description of the barrier.

In Fig. 3(d), we present histograms of resistance for junctions that have been grouped by the breakdown voltage of the junction, distinguishing junctions as being above or below the midpoint of the bimodal distribution, 1.3 V. We show a difference in average resistance of the high vs low breakdown junctions of $\sim 300 \Omega$, a $\sim 4\%$ change.

Here, we show how a fabrication process can be assessed considering breakdown voltages, where the distribution of breakdown voltages could be used as feedback when optimizing junction growth protocols, and correlations between breakdown voltages and resistances can be used to identify whether variation between barriers is contributing significantly to resistance spread. Specifically, we can identify two sub-ensembles of junctions within the nominally identical family of junctions that cannot be resolved from resistance measurements alone. As mentioned in the Introduction, it has recently been suggested that these thinnest points in junctions can contribute deviations of the $C\phi R$ from an ideal sinusoidal relationship, causing effects such as Josephson harmonics

[20,21]. This provides additional motivation to use this type of optimization protocol, as it may allow manufacturing processes to realize more homogeneous $C\phi R$ s.

In Fig. 3(e), we show the thickness distribution of two example simulated junctions. We find that the distributions overlap quite well for counts at thinner distributions. Given that the junctions have similar resistances, this is good evidence that the majority of the conductance is determined by the thinner portions of the barrier. We see this again in Fig. 3(f), where we plot a cumulative conductance distribution as a function of junction area and find a large contribution to total conductance coming from a small fraction of the total area, irrespective of whether using a skewed lognormal distribution or a normally distributed thickness. Areas of locally elevated conductance are more prominent in the normally distributed junction, as is to be expected. As the thicker regions contribute little to the total conductance, our model has bad resolution on the thicker side of the thickness distribution. The key feature of the lognormal probability distribution, that our modeling does provide confidence for, is that the tails on the thinner side of the distribution are suppressed more strongly than in the normal distribution. The observation of a small fraction of the junction contributing a large fraction of conductance agrees qualitatively with the conclusions from Refs. [41,45]. Both studies arrive at this conclusion from considerations of barrier thickness measurements by (S)TEM, which we consider in the next section.

III. SCANNING TRANSMISSION ELECTRON MICROSCOPY

STEM includes a family of techniques that allow for sub-nanometre resolution imaging of materials with elemental contrast, making them natural candidates to investigate questions about the thickness distribution of the barrier. These techniques can be applied to JJs where the barrier has been capped with a top electrode, i.e., after the full fabrication process has taken place. We first consider a protocol to measure variations in thickness in the projected 2D image. We do this considering images of the oxygen content in a barrier collected by STEM energy-dispersive x-ray spectroscopy (EDS) mapping. Imaging is performed on a lamella that is fabricated by focused ion beam cutting and lift out of a ~ 30 -nm-thick cross section of the JJ. In Fig. 4, we present an example image of the oxygen composition. The edge of the barrier is identified by kernel integration using OpenCV [46]. We define two kernels to identify the two edges, where each kernel has an asymmetry factor δ that differentially changes the weight of pixels on the left- and right-hand side of the kernel. We describe the construction of the integration kernels in the Supplemental Material [34]. Given that the oxygen content in the barrier increases over a finite length, the edge of the barrier could be defined at several points. By changing δ , we systematically alter where the kernel integration protocol identifies the edge. In Figs. 4(a)–4(c), we show the same image with edges identified using 0, 0.2, 0.4 as values of δ , respectively. In analysis, we explored larger values of δ and found the edges were obviously within the junction, so care must be taken in selecting this asymmetry value.

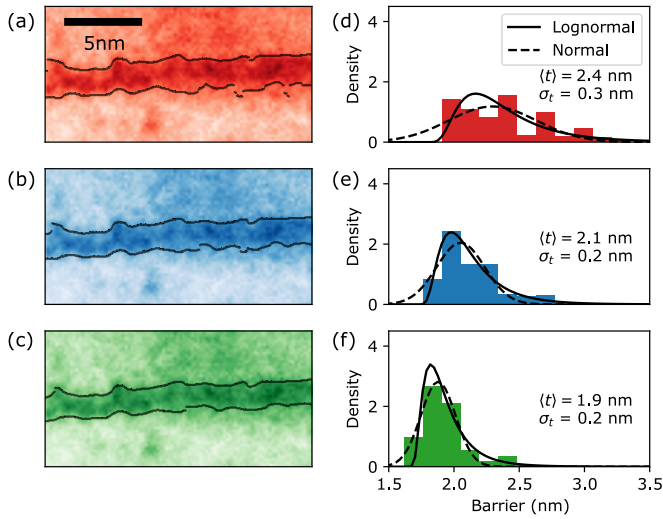


FIG. 4. Experimental STEM-EDS imaging of an Al/AIO_x/Al barrier showing the oxygen peak. (a)–(c) Show heatmaps indicating the oxygen peak with the edges of the barrier as detected by kernel integration overlaid on the maps. Different values of δ are used to define the kernel for the three maps (0, 0.2, 0.4, respectively). (d)–(f) Show histograms of thicknesses as inferred from the different kernel integrations.

We measure the thickness of the barrier by finding the distance between the two edges for each column of pixels. The average of the thickness from these different kernels matches previous measurements by TEM well, giving an average thickness of ~ 1.9 – 2.4 nm, depending on the kernel [26]. We can also compute the distribution of thicknesses shown in the histograms Figs. 4(d)–4(f) and fit each of them to both normal and lognormal distributions. We routinely see that the lognormal distribution fits the thickness distribution better than the normal distribution. In the literature, the results of this type of analysis have been interpreted as the statistical distribution of thicknesses of a barrier, a claim we are explicitly not making here.

Arbitrary choices, such as the asymmetry factor in the kernels, can change what we infer about the barrier. We attribute this both to (i) details of the barrier, such as the oxygen content likely changing over a finite distance and the nonuniform barrier thickness, and (ii) the STEM image being a projection that averages the barrier. As the barrier is grown on a bottom electrode that is not perfectly flat, the imaging beam is at times intersecting both electrode and barrier. We wish to understand (i) from techniques such as STEM imaging as they will determine the behavior of a JJ. However, (ii) represents difficulties inherent to trying to identify thickness variations in this material system by STEM. We suggest that we should report $1.9 \text{ nm} < \langle t \rangle < 2.4 \text{ nm}$, i.e., the variation in thickness implied by different kernels is what defines the error bars on the measurement of the average barrier thickness.

In order to gain a deeper understanding of (ii) from above, we simulate STEM EDS images of oxygen content. We simulate the STEM image by building a 3D array of 0.1 nm^3 voxels that represents the lamella used for STEM imaging. We use AFM maps to determine the topography of the barrier and then assign voxels in the barrier a value of 1, representing

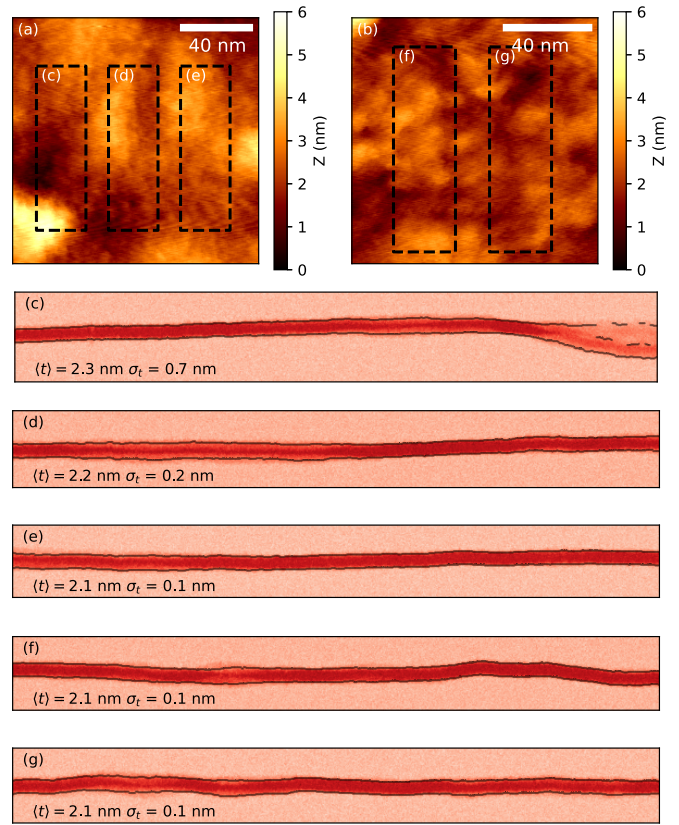


FIG. 5. (a), (b) Two high-resolution AFM scans of representative bottom leads [region indicated in Fig. 1(a)]. The root mean-squared roughness for the areas shown are 0.7 and 0.5 nm, respectively. In (a) and (b), five $30 \times 100 \text{ nm}$ areas are marked by dashed lines. These regions are used to simulate STEM profiles shown in (c)–(g) assuming a 2-nm oxide barrier coating the AFM topography, with the labels matching the AFM region to the STEM profile inserted into the boxes. (c)–(g) Edges of simulated oxygen content are detected using symmetric kernels and shown overlaid, as well as measurements of average thickness and thickness standard deviations noted in each figure. The labels (c)–(g) are in the same side of the scan region in the simulated STEM profile and the AFM regions.

high oxygen content, voxels on the edge of the barrier a value of 0.5 and other voxels a value of 0, representing low oxygen content. Averaging values of voxels along a column normal to the lamella surface gives a value corresponding to the nominal oxygen content in that column and represents the projection formed by the electron beam experimentally. After projection, we add Gaussian noise to the image, representative of detector noise, and calibrate the center and standard deviation of the Gaussian noise from a line trace of the experimental data in Fig. 4. We finally apply a Gaussian blur with radius 0.1 nm, representative of the beam size in STEM experiments.

We generate a series of barriers assuming that the thickness of the barrier is uniformly 2 nm across the junction. Using a uniform barrier thickness means that any thickness variation inferred from the simulated STEM image is fictitious, indicative of what we may expect to see from experimental nonidealities that could change what we infer from analysis of STEM images. We draw the bottom electrode topography from AFM measurements of the bottom lead shown in Figs. 5(a) and

5(b). These scans were taken from a region approximately overlapping the text “Bottom Lead” in Fig. 1(a) [47]. We infer the thickness of the barriers from the simulated STEM images [Fig. 5(c)–5(g)] using symmetric kernels ($\delta = 0$) as described for Fig. 4, adjusting kernel sizes to account for the different pixel sizes. As expected, a perfectly uniform barrier appears to have varying thickness when viewed in projection due to roughness in the underlying barrier. We also find that the thickness values and thickness standard deviation change depending on which AFM area is used to build the projection. All but one average thickness measurement is within 10% of the barrier thickness input to the simulation, although the standard deviation of barrier thickness varies by a factor of $\sim 7\times$. This suggests that STEM is a good technique to measure the average thickness of a barrier (reporting errors as described above), but calls into question the robustness of using STEM profiles to infer the thickness distribution in a junction. It also shows that multiple areas should be measured, and inferring average thickness values from single STEM images may introduce errors. We therefore include additional STEM images with analogous processing to that performed in Fig. 4 in the Supplemental Material [34]. These suggest similar thicknesses, but also include regions of topographic features, which slightly increase the inferred average thickness.

Sometimes it is straightforward to identify regions where apparent thickness broadening occurs because of topographic variation in the bottom electrode [e.g., right-hand side of Fig. 5(c)] where it is possible to resolve two barriers superimposed on one another. Here, the kernel edge detection does not work well, shown by the discontinuities in the edge it identifies. In the Supplemental Material [34] we show an example of experimental STEM imaging with these types of features, compared to a similar STEM simulation. In Fig. 5(d), there is also a topographic gradient along the projection axis, but, in this instance, the topographic change is smaller in magnitude and so, in the simulated STEM image, it appears only as a broadening of the barrier. It is therefore not straightforwardly possible to systematically distinguish apparent broadening from topography variation and true barrier variation, although it is possible to exclude areas where the topographic broadening effects are largest.

Comparing experimental (Fig. 4) and simulated (Fig. 5) STEM images, barriers appear rougher in the experimental STEM data than in the simulated images. The topography of the electrode used to simulate STEM images may be artificially smoothed because of the AFM tip radius of 2 nm. This would mean that features on the scale of the apparent roughness seen in Fig. 4 would not be apparent with this AFM tip. The barriers we measure here are also oxidized, which can change the topography of the surface. While there may be features sufficiently small that they are not resolved by AFM, both AFM maps in Fig. 5 have topographic features that are large relative to the AFM resolution, and, as we show, couple importantly into the inferred STEM projections. Using STM may allow higher-resolution topographic maps to perform more robust inferences of STEM profiles, and can also be performed *in situ* before barrier oxidation modifies the topography. These more detailed maps might find uses, for instance, in reverse Monte Carlo approaches [48] to

extract quantitative information about the barrier thickness and thickness distributions from STEM.

IV. CONCLUSIONS

In this article, we have aimed to understand oxide barriers in JJs because of their technological importance in superconducting qubits. A summary of different thicknesses inferred within this paper is shown in Table I. We fit the Simmons model to nonlinear IVs to extract a single-valued thickness value to each junction ~ 0.78 nm. We build Monte Carlo protocols to simulate the IV properties of barriers with multivalued thicknesses drawn from representative statistical distributions. Here, we find that the distribution of thicknesses present in the junction, results in IVs that, when fitted, would imply a single-valued thickness smaller than the average thickness. This is because conductivity is exponentially dependent on the barrier thickness, so any thinner-than-average regions contribute more than equally thicker-than-average regions. These simulations indicate a barrier that is ~ 1 -nm thick with a standard deviation of ~ 0.2 -nm; however, using different barrier heights, we infer different thickness distributions. The rectangular potential barrier used in the Simmons model likely results in an underestimate of thickness relative to more realistic, smoothly increasing barriers. By considering the Monte Carlo simulations relative to STEM measurements, we constrain the barrier height to the range $0.8 \text{ eV} \lesssim \phi \lesssim 1.5 \text{ eV}$ with best matching at $\sim 1.2 \text{ eV}$.

Using breakdown measurements we can probe the tails of thickness distributions assuming that breakdown occurs at the thinnest point or points of the barrier. Considering normal and skewed lognormal distributions of barrier thickness, we show that the breakdown statistics are better described by a skewed distribution. We show that the tails of the thickness distribution contribute substantially to the resistance of the device, with a $\sim 4\%$ difference in resistance between the 93% lowest- and 7% highest-breakdown voltage junctions. We also show that normally distributed barrier thicknesses give rise to larger barrier-induced variations than skewed distributions.

We then consider STEM measurements probing the oxygen content of barriers as a technique to directly measure the thickness of barriers. We show measurements of the barrier thickness from 1.9 to 2.4 nm depending on how the edge of the barrier is determined, with a standard deviation not dissimilar to those inferred from the Monte Carlo simulations of the barrier. We suggest an approach to define errors on barrier thickness measurements, performing kernel integration with different kernels to extract different barrier edges and comparing the variation in the average thickness reported. These error bars should not be misinterpreted as the standard deviation of barrier thickness.

Comparing experimental measurements to simulations of STEM data, we show that inferring average thicknesses from STEM images is reasonable, subject to errors associated with identifying the edge of the barrier. However, inferring variations in the thickness of the barrier from STEM images is not a robust process. The roughness commonly seen on bottom electrodes, when projected into 2D, also creates apparent thickness variations. We see wider variations in barrier thickness measurements experimentally than when simulating

TABLE I. A summary of different barrier thicknesses extracted throughout this paper. Fitting the Simmons model returns a single-valued thickness and the error represents the standard deviation from fitting 597 IVs. Monte Carlo simulations give thickness by fixing the area of a junction and allowing the barrier thickness to vary. The thickness value is the average thickness whereas the errors represent a genuine thickness distribution in the barrier. STEM-EDS measurements are direct measurements of thickness by atomic scale microscopy. As shown here, the thickness of the barrier is sensitive to image processing (i.e., choice of δ). Errors here represent standard deviations of inferred average thickness for a specific choice of δ but do not represent thickness distributions. Without an unbiased approach to image processing, the true error on thickness measurements is likely the variation in thickness between different values of δ .

Technique	Simmons model fit	Monte Carlo simulations				STEM-EDS		
		Potential barrier height (eV) only lognormal distribution				Kernel asymmetry (δ)		
Variable		0.8	1.0	1.22	1.5	0	0.2	0.4
Thickness (nm)	0.78 ± 0.02	No fit	1.3 ± 0.3	1.0 ± 0.2	0.8 ± 0.1	2.4 ± 0.3	2.1 ± 0.2	1.9 ± 0.2

a perfectly uniform barrier, lending support to the idea that the barrier thickness does indeed vary across the junction but inferring the statistics of this variation from STEM data alone remains a challenge. Most evidence in the literature about thickness distributions of barriers being normally distributed arises from this type of processing of STEM or TEM data, for instance [26]. While these works were substantial contributions to the understanding of oxide barriers in JJs, continuing to use these techniques and inferences should be avoided, for instance, in computations of Josephson harmonics based on normal thickness distributions [21]. We show here that this type of inference overstates the certainty of thickness distributions, and indeed our most sensitive probe of the tails of our distribution implies some form of skewed thickness distribution, contradicting the conclusions drawn from STEM.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available because they contain commercially sensitive information. The data are available from the authors upon reasonable request.

- [1] F. Arute, K. Arya, R. Babbush, D. Bacon, J. C. Bardin, R. Barends, R. Biswas, S. Boixo, F. G. Brandao, D. A. Buell *et al.*, Quantum supremacy using a programmable superconducting processor, *Nature (London)* **574**, 505 (2019).
- [2] C. Macklin, K. O'Brien, D. Hover, M. Schwartz, V. Bolkhovsky, X. Zhang, W. Oliver, and I. Siddiqi, A near-quantum-limited Josephson traveling-wave parametric amplifier, *Science* **350**, 307 (2015).
- [3] S. Nik, P. Krantz, L. Zeng, T. Greibe, H. Pettersson, S. Gustafsson, P. Delsing, and E. Olsson, Correlation between Al grain size, grain boundary grooves and local variations in oxide barrier thickness of Al/AIO_x/Al tunnel junctions by transmission electron microscopy, *SpringerPlus* **5**, 1067 (2016).
- [4] S. Fritz, L. Radtke, R. Schneider, M. Luysberg, M. Weides, and D. Gerthsen, Structural and nanochemical properties of AIO_x layers in Al/AIO_x/Al-layer systems for Josephson junctions, *Phys. Rev. Materials* **3**, 114805 (2019).
- [5] L. Zeng, D. T. Tran, C.-W. Tai, G. Svensson, and E. Olsson, Atomic structure and oxygen deficiency of the ultrathin aluminium oxide barrier in Al/AIO_x/Al Josephson junctions, *Sci. Rep.* **6**, 29679 (2016).
- [6] P. Lapham and V. P. Georgiev, Computational study of oxide stoichiometry and variability in the Al/AIO_x/Al tunnel junction, *Nanotechnology* **33**, 265201 (2022).
- [7] D. Yohannes, S. Sarwana, S. K. Tolpygo, A. Sahu, Y. A. Polyakov, and V. K. Semenov, Characterization of HYPRES' 4.5 kA/cm² & 8 kA/cm² Nb/AIO_x/Nb fabrication processes, *IEEE Trans. Appl. Supercond.* **15**, 90 (2005).
- [8] J. T. West, A. Kurler, A. Wynn, C. Rogers, M. A. Gouker, and S. K. Tolpygo, Wafer-scale characterization of a superconductor integrated circuit fabrication process, using a cryogenic wafer prober, *IEEE Trans. Appl. Supercond.* **32**, 1 (2022).
- [9] V. K. Semenov, Y. A. Polyakov, and S. K. Tolpygo, Very large scale integration of Josephson-junction-based superconductor random access memories, *IEEE Trans. Appl. Supercond.* **29**, 1 (2019).
- [10] J. Kreikebaum, K. O'Brien, A. Morvan, and I. Siddiqi, Improving wafer-scale Josephson junction resistance variation in superconducting quantum coherent circuits, *Supercond. Sci. Technol.* **33**, 06LT02 (2020).
- [11] V. Ambegaokar and A. Baratoff, Tunneling between superconductors, *Phys. Rev. Lett.* **10**, 486 (1963).
- [12] A. Osman, J. Simon, A. Bengtsson, S. Kosen, P. Krantz, D. P. Lozano, M. Scigliuzzo, P. Delsing, J. Bylander, and A. Fadavi Roudsari, Simplified Josephson-junction fabrication process for reproducibly high-performance superconducting qubits, *Appl. Phys. Lett.* **118**, 064002 (2021).
- [13] D. O. Moskalev, E. V. Zikiy, A. A. Pishchimova, D. A. Ezenkova, N. S. Smirnov, A. I. Ivanov, N. D. Korshakov, and

- I. A. Rodionov, Optimization of shadow evaporation and oxidation for reproducible quantum Josephson junction circuits, *Sci. Rep.* **13**, 4174 (2023).
- [14] A. A. Pishchimova, N. S. Smirnov, D. A. Ezenkova, E. A. Krivko, E. V. Zikiy, D. O. Moskalev, A. I. Ivanov, N. D. Korshakov, and I. A. Rodionov, Improving Josephson junction reproducibility for superconducting quantum circuits: Junction area fluctuation, *Sci. Rep.* **13**, 6772 (2023).
- [15] A. Osman, J. Fernández-Pendás, C. Warren, S. Kosen, M. Scigliuzzo, A. Frisk Kockum, G. Tancredi, A. Fadavi Roudsari, and J. Bylander, Mitigation of frequency collisions in superconducting quantum processors, *Phys. Rev. Res.* **5**, 043001 (2023).
- [16] N. Acharya, R. Armstrong, Y. Balaji, K. G. Crawford, J. C. Gates, P. C. Gow, O. W. Kennedy, R. D. Pothuraju, K. Shahbazi, and C. D. Shelly, Integration of through-sapphire substrate machining with superconducting quantum processors, *Adv. Mater.* **37**, 2411780 (2024).
- [17] J. Van Damme, S. Massar, R. Acharya, T. Ivanov, D. Perez Lozano, Y. Canvel, M. Demarets, D. Vangoidsenhoven, Y. Hermans, J. Lai *et al.*, Advanced CMOS manufacturing of superconducting qubits on 300 mm wafers, *Nature (London)* **634**, 74 (2024).
- [18] B. D. Josephson, Possible new effects in superconductive tunnelling, *Phys. Lett.* **1**, 251 (1962).
- [19] A. A. Golubov, M. Y. Kupriyanov, and E. Il'Ichev, The current-phase relation in Josephson junctions, *Rev. Mod. Phys.* **76**, 411 (2004).
- [20] K. Bayros, M. J. Cyster, J. S. Smith, and J. H. Cole, Influence of pinholes and weak-points in aluminum-oxide Josephson junctions, *Phys. Rev. Materials* **8**, 046202 (2024).
- [21] D. Willsch, D. Rieger, P. Winkel, M. Willsch, C. Dickel, J. Krause, Y. Ando, R. Lescanne, Z. Leghtas, N. T. Bronn *et al.*, Observation of Josephson harmonics in tunnel junctions, *Nat. Phys.* **20**, 815 (2024).
- [22] B. H. Lee, J. Oh, H. H. Tseng, R. Jammy, and H. Huff, Gate stack technology for nanoscale devices, *Mater. Today* **9**, 32 (2006).
- [23] D. Buchanan and S.-H. Lo, Reliability and integration of ultra-thin gate dielectrics for advanced CMOS, *Microelectron. Eng.* **36**, 13 (1997).
- [24] S. R. Ekanayake, M. Ford, and M. Cortie, Metal-insulator-metal (MIM) nanocapacitors and effects of material properties on their operation, *Mater. Forum* **27**, 15 (2004).
- [25] V. Da Costa, C. Tiusan, T. Dimopoulos, and K. Ounadjela, Tunneling phenomena as a probe to investigate atomic scale fluctuations in metal/oxide/metal magnetic tunnel junctions, *Phys. Rev. Lett.* **85**, 876 (2000).
- [26] L. Zeng, S. Nik, T. Greibe, P. Krantz, C. Wilson, P. Delsing, and E. Olsson, Direct observation of the thickness distribution of ultra thin AlO_x barriers in $\text{Al}/\text{AlO}_x/\text{Al}$ Josephson junctions, *J. Phys. D* **48**, 395308 (2015).
- [27] E. R. Dobrovinskaya, L. A. Lytvynov, and V. Pishchik, Properties of sapphire, in *Sapphire: Material, Manufacturing, Applications* (Springer, Boston, MA, 2009), pp. 55–176.
- [28] X. Liu, K. Pan, Z. Zhang, and Z. Feng, Unveiling atomic structure and chemical composition of the $\text{Al}/\text{AlO}_x/\text{Al}$ Josephson junctions in qubits, *Appl. Surf. Sci.* **640**, 158337 (2023).
- [29] E. Supple, M. Holtz, C. J. Richardson, and B. Gorman, Atomic structure of superconducting tunnel junctions using STEM and APT, *Microsc. Microanal.* **27**, 2460 (2021).
- [30] M. Cyster, J. Smith, N. Vogt, G. Opletal, S. Russo, and J. Cole, Simulating the fabrication of aluminium oxide tunnel junctions, *npj Quantum Inf.* **7**, 12 (2021).
- [31] F. Palumbo, C. Wen, S. Lombardo, S. Pazos, F. Aguirre, M. Eizenberg, F. Hui, and M. Lanza, A review on dielectric breakdown in thin dielectrics: Silicon dioxide, high- k , and layered dielectrics, *Adv. Funct. Mater.* **30**, 1900657 (2020).
- [32] G. Dolan, Offset masks for lift-off photoprocessing, *Appl. Phys. Lett.* **31**, 337 (1977).
- [33] J. G. Simmons, Generalized formula for the electric tunnel effect between similar electrodes separated by a thin insulating film, *J. Appl. Phys.* **34**, 1793 (1963).
- [34] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/5tb4-pslg> for (I) discussion of the Simmons fitting to IVs, (II) AFM measurements of typical junction dimensions, (III) additional STEM imaging of other barrier regions, (IV) spatial maps of barrier height and thickness from Simmons models as well as breakdown voltages, (V) discussion of the kernel integration protocol used to detect barrier edges, and (VI) additional Monte Carlo simulations.
- [35] M. Koberidze, A. Feshchenko, M. Puska, R. Nieminen, and J. Pekola, Effect of interface geometry on electron tunnelling in $\text{Al}/\text{Al}_2\text{O}_3/\text{Al}$ junctions, *J. Phys. D* **49**, 165303 (2016).
- [36] M. Koberidze, M. J. Puska, and R. M. Nieminen, Structural details of $\text{Al}/\text{Al}_2\text{O}_3$ junctions and their role in the formation of electron tunnel barriers, *Phys. Rev. B* **97**, 195406 (2018).
- [37] L. Dorneles, D. Schaefer, M. Carara, and L. Schelp, The use of Simmons' equation to quantify the insulating barrier parameters in $\text{Al}/\text{AlO}_x/\text{Al}$ tunnel junctions, *Appl. Phys. Lett.* **82**, 2832 (2003).
- [38] C.-E. Kim, K. G. Ray, and V. Lordi, A density-functional theory study of the $\text{Al}/\text{AlO}_x/\text{Al}$ tunnel junction, *J. Appl. Phys.* **128**, 155102 (2020).
- [39] P. Snijders, L. Jeurgens, and W. Sloof, Structure of thin aluminium-oxide films determined from valence band spectra measured using XPS, *Surf. Sci.* **496**, 97 (2002).
- [40] S. Fritz, L. Radtke, R. Schneider, M. Weides, and D. Gerthsen, Optimization of $\text{Al}/\text{AlO}_x/\text{Al}$ -layer systems for Josephson junctions from a microstructure point of view, *J. Appl. Phys.* **125**, 165301 (2019).
- [41] J.-S. Oh, C. J. Kopas, H. Cansizoglu, J. Y. Mutus, K. Yadavalli, T.-H. Kim, M. Kramer, A. H. King, and L. Zhou, Correlating aluminum layer deposition rates, Josephson junction microstructure, and superconducting qubits' performance, *Acta Mater.* **284**, 120631 (2025).
- [42] J. Strand and A. L. Shluger, On the structure of oxygen deficient amorphous oxide films, *Adv. Sci.* **11**, 2306243 (2024).
- [43] 0.2 nm approximates the ionic radii in the AlO_x barrier.
- [44] J. Kolodzey, E. A. Chowdhury, T. N. Adam, G. Qui, I. Rau, J. O. Olowolafe, J. S. Suehle, and Y. Chen, Electrical conduction and dielectric breakdown in aluminum oxide insulators on silicon, *IEEE Trans. Electron Devices* **47**, 121 (2000).
- [45] T. Aref, A. Averin, S. van Dijken, A. Ferring, M. Koberidze, V. F. Maisi, H. Nguyend, R. M. Nieminen, J. P. Pekola, and L. Yao, Characterization of aluminum oxide tunnel barriers by combining transport measurements and transmission electron microscopy imaging, *J. Appl. Phys.* **116**, 073702 (2014).
- [46] G. Bradski, The OpenCV Library, Dr. Dobb's Journal of Software Tools (2000), <https://github.com/opencv/opencv/wiki/CiteOpenCV>.

- [47] Noise at the ~ 0.6 -nm range is present in some regions of these AFM scans, which we found hard to exclude from measurements. This represents $\sim 10\%$ of the total topographic variation in these AFM maps and is not present across the whole scans and therefore we believe not a dominant contributor to the following analysis.
- [48] R. L. McGreevy, Reverse Monte Carlo modelling, *J. Phys.: Condens. Matter* **13**, R877 (2001).