

Fabrication and Structural Analysis of Trilayers for Tantalum Josephson Junctions with Ta₂O₅ Barriers

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Tantalum (Ta) has recently emerged as a promising low-loss material, enabling record coherence times in superconducting qubits. This enhanced performance is largely attributed to its stable native oxide, which is believed to host fewer two-level system (TLS) defects – key contributors to decoherence in superconducting circuits. Nevertheless, aluminum oxide (AlO_x) remains the predominant choice for Josephson junction barriers in most qubit architectures. In this study, we systematically investigate various techniques for forming high-quality oxide layers on α -phase tantalum (α -Ta) thin films, aiming to develop effective Josephson junction barriers. We explore thermal oxidation in a tube furnace, rapid thermal annealing, as well as plasma oxidation of both room-temperature and heated Ta films, and propose a mechanistic picture of the underlying oxidation mechanisms. All methods yield Ta₂O₅, the same compound as tantalum’s native oxide. Among these, plasma oxidation produces the smoothest and highest-quality oxide layers, making it particularly well-suited for Josephson junction fabrication. Furthermore, we demonstrate the successful epitaxial growth of α -Ta atop oxidized α -Ta films, paving the way for the realization of trilayer Ta/Ta-O/Ta Josephson junctions with clean, low-loss interfaces.

I. INTRODUCTION

Superconducting circuits designed to operate as artificial atoms, exhibiting discrete energy levels, are a promising platform for scalable quantum computing. These micron-sized circuits combine inductive and capacitive components with nanoscale Josephson junctions — two superconductors sandwiching a non-superconducting barrier layer. Josephson junctions induce a non-linear response such that the two lowest energy states can be selectively addressed. Information is then encoded in the charge, flux, or phase of the circuit and stored for time frames limited by decoherence.¹ Although the precise microscopic origins of decoherence in Josephson junctions are not completely understood, they can be traced to defects in the superconductor, the tunneling barrier, and at the interfaces. These defects become problematic if they form two-level systems (TLSs), which have dipole moments that interact with operating currents, resulting in energy relaxation and dephasing.^{2–5}

Aluminum is often considered the silicon of superconducting qubits. Most fabrication processes take advantage of aluminum’s self-terminating native oxide to create an amorphous tunnel barrier in aluminum-based Josephson junctions.⁶ However, this native oxide is known to be a significant source of defects that contribute to decoherence, leading researchers to explore alternative materials.^{5,7–10} Notably, tantalum (Ta) has recently emerged as a promising material for low-loss quantum circuits. Tantalum resonators have demonstrated high quality factors up to 15×10^6 in the single-photon regime,^{11–13} exceeding the highest values observed in aluminum resonators.¹⁴

Moreover, tantalum-based qubits have achieved record lifetimes (T_1) above 0.5 ms,^{15,16} a significant improvement over aluminum-based qubits.¹⁷ However, these Ta-based qubits still depend on lossy AlO_x junction barriers.

The native oxide of tantalum (TaO_x) is thinner, forms cleaner interfaces,¹⁸ and may contain fewer TLS defects than that of niobium (Nb), another commonly-used material in superconducting circuits.¹⁹ In particular, the tantalum’s native oxide exhibits a closer-to-crystalline bonding nature with less disorder than that of Nb, likely mitigating the creation of TLSs by limiting atomic hydrogen diffusion to the interface.²⁰ The stability of the oxide has also been demonstrated through high quality factors in tantalum resonators measured over several months.²¹ Given these favorable characteristics, it is compelling to investigate whether tantalum-based Josephson junctions—featuring tantalum-oxide barriers—can achieve reduced losses. This exploration necessitates establishing a reliable growth method for tantalum-oxide and trilayer structures in which superconducting α -phase tantalum encloses the tantalum-oxide barrier.

In this study, we test three protocols for creating a tantalum-oxide layer for Ta/Ta-O/Ta Josephson junctions — tube furnace annealing, rapid thermal annealing, and oxygen plasma annealing in a sputtering system — in each case varying the annealing temperature and time. We then measure the oxide thicknesses using X-ray reflectometry (XRR), characterize oxide species using X-ray photoelectron spectroscopy (XPS), analyze grain structure using atomic force microscopy (AFM) and electron microscopy, and construct the mechanistic picture of the oxidation process using ab initio modeling based on the density functional theory (DFT). Lastly, we test different processes for

growing α -phase Ta on top of the oxidized Ta layer, to form a trilayer structure necessary for a Ta-based Josephson junction.

II. EXPERIMENTAL DETAILS

Crystalline Ta exists in two phases: the alpha phase (α -Ta), which has a body-centered cubic (BCC) structure, and the beta phase (β -Ta), which has a tetragonal crystalline structure and is metastable.^{22,23} Because α -Ta has a superconducting critical temperature T_c that is substantially higher than that of β -Ta (4.4 K versus 0.6-1 K), α -Ta is preferred for superconducting circuit applications.

To promote the growth of α -Ta, we deposited Ta via DC magnetron sputtering using two different procedures: high-temperature growth on sapphire substrates and room-temperature growth on silicon substrates using a niobium (Nb) seed layer.²⁴⁻²⁶ In the former, 150 nm of Ta was deposited on sapphire substrates held at 500 °C, as substrate heating above 400 °C is known to result in the growth of pure α -Ta.²⁷ In the latter, 60 or 100 nm (see Table I) of Ta was deposited on Si(004) substrates atop a 6-nm Nb seed layer. Further details regarding the growth process are included in the Methods section. Lastly, we verified that we obtained α -Ta using X-ray diffraction and measurements of a $T_c \approx 4.3$ -4.4 K using magnetometry, as shown in Supplementary Fig. S1.

To develop a protocol for creating a tantalum-oxide barrier for fully tantalum-based Josephson junctions, we tested three different oxidation procedures: annealing Ta films in (1) an Expertech CTR200 tube furnace under a flow of oxygen, (2) an oxygen flow inside a AccuThermo AW 610 rapid thermal processing system, and (3) an oxygen plasma in a Lesker Lab 18 sputtering chamber. Table I summarizes the procedures applied to each sample.

First, the tube furnace annealing process involved heating the system to 350 °C at a rate of 10 °C/min then at 2.5 °C/min until the final temperature reached 400 °C. To prevent any oxidation during the ramp-up process, the furnace was flushed with nitrogen (N_2) gas. Upon achieving 400 °C, the system was flushed with O_2 at a rate of 20,000 sccm for 10-60 minutes (see Table I). Immediately following the annealing process, the heater was turned off, and the system was allowed to cool under a continuous flow of nitrogen gas.

Secondly, to test a more accelerated oxidation process, we employed rapid thermal annealing, taking advantage of its swift temperature ramp-up and ramp-down capabilities. The films were heated to 700 °C at a rate of 70 °C/sec under a 5,000 sccm flow of O_2 and held for durations ranging from 1 to 10 minutes, as detailed in Table I. As with the tube furnace annealing method, the system was purged with N_2 during all other stages and subsequently cooled to room temperature following the annealing step.

Finally, for plasma oxidation, the films were exposed to a 100 W RF plasma in a sputtering chamber, using a 20 sccm flow of oxygen gas. The base pressure of the chamber prior to oxidation was less than 6×10^{-8} Torr. Oxidation was then carried out at room temperature, as well as with the substrate heated to 200 °C, 300 °C, and 400 °C to assess temperature-

dependent effects.

III. RESULTS AND DISCUSSION

When a non-superconducting barrier separates two superconducting electrodes, the superconductors' order parameters decay exponentially into the barrier. For a barrier thickness d that is sufficiently thin compared to the superconductor coherence length (ξ), the order parameters overlap. This overlap enables phase coherence between superconductors and a supercurrent of superconducting electron pairs (Cooper pairs) to flow across the barrier. In a superconductor-insulator-superconductor (SIS) junction, Cooper pair tunneling dominates when the barrier thickness $d < \xi$, whereas quasiparticle tunneling dominates for thicker barriers, resulting in lower supercurrent.²⁸

In epitaxial Ta films that exhibit 2D superconductivity, the in-plane coherence length of α -Ta has been measured to vary between 21.7 nm and 54.8 nm, for films up to 162.2 nm thick.²⁹ These values set an effective upper limit to the thickness of an insulating barrier for SIS tunnel junctions based on Ta. Consequently, we must develop a process that can produce thin, fully oxidized barriers without significant inter-metallic Ta that could form a superconducting short between electrodes. Moreover, it is important to identify the type of oxide formed for consideration of the potential sources of energy loss.

A. Oxygen Species

To determine the chemical state of the oxidized layer in our films, we performed X-ray photoelectron spectroscopy, a technique that employs the photoelectric effect to obtain the chemical composition of the top layer of a material. We used a Kratos Axis-Ultra DLD spectrometer equipped with an Al $K\alpha$ X-ray source operating at 15 kV and 225 W. For these acquisitions, the X-ray spot size was approximately $700 \times 300 \mu m^2$. Note that this spectrometer has a sample probe depth of 10 nm.

Figure 1 shows the high-resolution photoelectron Ta(4f) spectra for all films. First, focusing on the control film that only contains a native surface oxide, we observe two pairs of spin-orbit split double peaks, corresponding to the expected peak positions of the Ta metal doublet and Ta_2O_5 doublet.^{30,31} Specifically, we identify the peaks at binding energies of 26.8 eV and 28.8 eV as the Ta $4f_{7/2}$ and $4f_{5/2}$ valence states of the Ta^{5+} ion in stoichiometric Ta_2O_5 . The peaks at lower binding energies of 21.6 eV and 23.6 eV are consistent with expectations for the $4f_{7/2}$ and $4f_{5/2}$ valence states of metallic tantalum Ta^0 . These results are consistent with other XPS studies on Ta.^{15,32-36} We observe a similar peak structure in the spectra collected from the two Ta films exposed to an oxygen plasma at room temperature (samples 9, 10). The spectra produced by the remaining films exhibit only the Ta_2O_5 doublet, suggesting complete oxidation within the top 10 nm of the film.

The slight binding energy shifts (0.7-0.8 eV range) observed in the Ta^{5+} 4f XPS spectra between samples result from

TABLE I: Summary of sample properties and results, including the substrate, Ta thickness (d_{Ta}), oxygenation process temperature (temp.), oxygen flow rate, oxygen annealing time, X-ray reflectometry, and atomic force microscopy results for oxidized samples. All samples on Si substrates include a sputtered 6-nm Nb seed layer below the Ta layer. Plasma oxidation was performed with 100 W of RF power biasing the substrate.

ID	Substrate	d_{Ta} [nm]	Oxygenation Process				TaO _x thickness [nm]	AFM roughness [nm]
			Method	Temp. [°C]	O ₂ [sccm]	Time [min]		
1	Si	100	—	22	—	—	1.93	0.518
2	Al ₂ O ₃	150	—	22	—	—	2.38	0.72
3	Si	100	Tube Furnace	400	20,000	10	37.14	1.04
4						30	56.66	1.24
5						60	60.32	1.71
6	Si	100	RTA	700	5,000	1	—	7.28
7						5	—	16.38
8						10	—	15.7
9	Si	60	Plasma	21.3	20	10	7.64	0.572
10		60		21.3		30	7.33	0.613
11		100		200		60	7.46	0.108
12		100		200		120	7.78	0.105
13		100		300		60	10.35	0.463
14		100		300		120	10.78	0.125
15		100		400		60	15.21	0.342
16		100		400		120	14.88	0.232

temperature-dependent oxygen vacancy formation. Different annealing temperatures create varying concentrations of oxygen vacancies in Ta₂O₅, with the formation of surface oxygen vacancies resulting in altered local electronic environments,³⁷ while oxygen gas availability during processing contributes to reducing oxygen vacancy density.³⁸ These process-induced variations in defect density create Ta₂O₅ layers with different electrical properties, leading to different degrees of surface charge buildup during XPS measurement, which shifts photoelectron binding energies.³⁹

B. Oxide Thickness

To determine the oxide thicknesses, we employed X-ray reflectometry using a Bruker D8 Discover with a Cu anode X-ray source (50kV, 1000 μ A). Figure 2 displays the measured XRR scattering intensities plotted against Q , the momentum transfer vector component perpendicular to the film surface. The solid curves are fits based on the Levenberg-Marquardt model, considering Ta₂O₅ and Ta layers, as well as the Nb seed layer (when applicable), and appropriate substrate. Note that the films oxidized using a rapid thermal annealing process were too rough for XRR; atomic force microscopy data determining the roughness is shown in section III C.⁴⁰

Based on the XRR fits shown in Fig. 2, the thickness of the oxide was extracted and summarized in Table I. For our native oxide samples, we extract a thickness of 1.93 and 2.38 nm, which is consistent with previous studies.^{18,41} For the tube furnace annealed films, XRR results show that the thickness of the oxide layer increased from 37 to 60 nm as the annealing time was increased from 10 to 60 min.

For the plasma oxidized films, we consider the effects of

different annealing temperatures as well as annealing times. First, the XRR measurements revealed that exposing the films to oxygen plasma on an unheated stage produced a surface oxide layer of approximately 7–8 nm thick. The extracted thickness for 10 minutes (sample 9) versus 30 minutes (sample 10) of exposure resulted in oxide thicknesses within 0.3 nm, suggesting that the process is insensitive to time within the tested time frames.

To test methods for generating a thicker oxide layer, we further exposed the samples to oxygen plasma while heating them for durations of 1 hour and 2 hours. Plasma oxidation at substrate temperatures of 200 °C, 300 °C, and 400 °C yielded oxide thicknesses of approximately 7–8 nm, 10 nm, and 15 nm, respectively (SEM cross-sectional images in Supplementary Fig. S2). Notably, the extra hour of the oxidation time had negligible influence on the final thickness in each case.

Figure 2(b) displays the resulting oxide thickness versus annealing temperature. The observed temperature-dependent oxide thickness in our plasma-oxidized samples can be explained by the thermodynamically controlled nature of tantalum oxidation. Ref. [42] used scanning transmission electron microscopy (STEM), electron energy-loss spectroscopy (EELS), and density functional theory (DFT) calculations to characterize tantalum films oxidized in air, revealing a three-layer structure: amorphous Ta₂O₅, crystalline TaO_{1- δ} interface, and unoxidized crystalline Ta. Their analysis showed that oxidation follows a defined pathway in which oxygen penetrates between Ta atomic planes, transitioning from ordered to amorphous structure when the O/Ta ratio exceeds 1:1. The time-independent behavior in our study suggests that each plasma oxidation temperature provides sufficient thermal energy to drive this structural reorganization to completion within 1 hour, with an increase in temperature increasing the effective depth

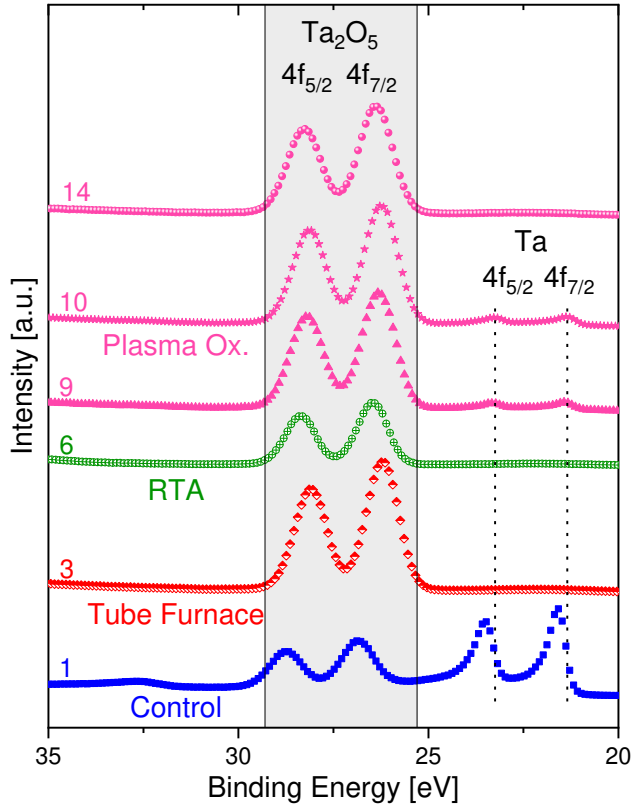


FIG. 1: High-resolution Ta(4f) XPS spectra of tantalum films – featuring a native oxide (blue curve, sample 1), as well as after tube furnace oxidation (red curve, samples 3), rapid thermal annealing (green curve, samples 6), and plasma oxidation (pink curves, samples 9,10,14) – reveal the characteristic Ta₂O₅ doublet peaks at binding energies of approximately 26.2 eV (Ta 4f_{7/2}) and 28.1 eV (Ta 4f_{5/2}). For some samples, peaks corresponding to metallic tantalum appear near 21.4 and 23.3 eV due to XPS probing deeper than the oxide thickness. Dotted lines mark the positions of the metallic Ta peaks, which are notably weak in plasma-oxidized samples 9 and 10.

of the O penetration. The final oxide thickness represents the thermodynamic equilibrium state for each temperature, explaining why extending the oxidation time beyond 1 hour does not increase thickness – the system has already reached its most stable configuration.

C. Surface Morphology

Superconducting quantum devices with rougher film surfaces may have higher losses than those of smoother films. For example, in one study,⁴³ reducing the RMS surface roughness of Nb-based superconducting resonators from 0.98 nm to 0.31 nm quintupled the internal quality factor Q_i . This was attributed to a reduction in losses from quasiparticles and TLSs. To quantify the surface roughness of our oxidized Ta films, we performed atomic force microscopy (AFM) over $1 \times 1 \mu\text{m}^2$ and $10 \times 10 \mu\text{m}^2$ scan areas. Further details are included in the

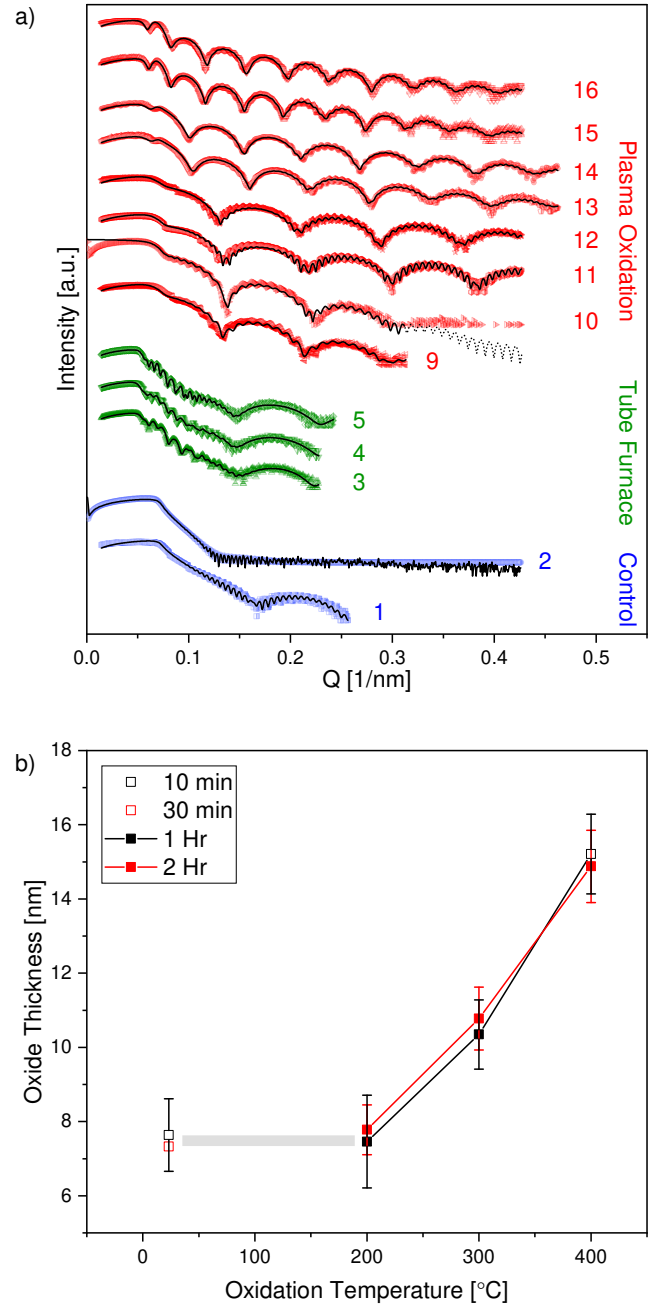


FIG. 2: (a) XRR measurements of Ta films subjected to native oxidation (control), tube furnace oxidation, and plasma oxidation treatments. Solid black lines show fitted curves used to extract the oxide thicknesses. The dotted portion of the sample 10 fit curve indicates poor fitting quality at high Q values. (b) Oxidation temperature vs. Thickness. A clear trend of approximately equal thickness can be seen for different temperatures and times. The gray line is a guide for the eye.

Methods section. Figure 3 displays the AFM images and Table I summarizes the mean roughness extracted from the $10 \mu\text{m}^2$ scans.

The Ta control film grown on a sapphire substrate at high temperature (sample 2) displays three primary domain orienta-

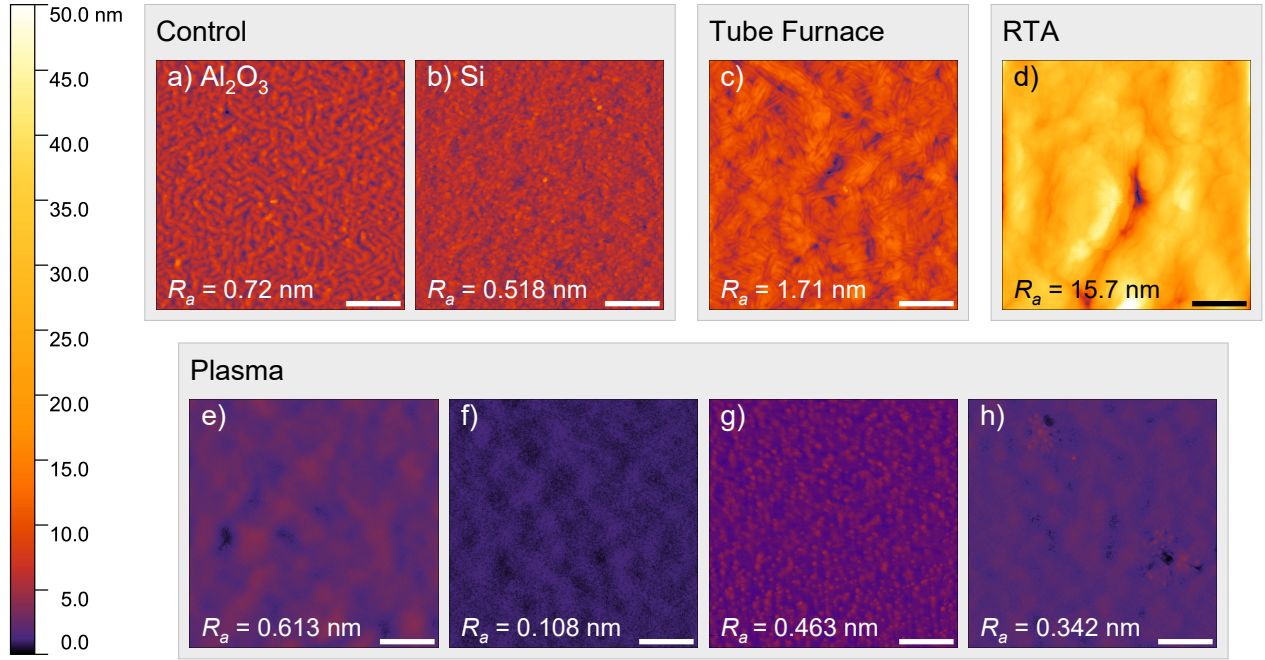


FIG. 3: Surface morphology of native and oxidized tantalum films characterized by AFM, with mean roughness (R_a) annotated for each sample. Tantalum films grown on (a) sapphire at 500 °C (sample 2) and (b) silicon at room temperature (sample 1) display distinct grain structures. (c) Thermal oxidation (sample 5) resulted in an oxide with large grain sizes and moderate surface roughness. (d) Rapid thermal annealing (sample 8) resulted in films with high surface roughness across a $10 \mu\text{m}^2$ scan region. Note that the pictured $1 \mu\text{m}^2$ scan region displays a lower roughness locally. Films that underwent plasma oxidation at (e) room temperature (sample 10), (f) 200 °C (sample 11), (g) 300 °C (sample 13), and (h) 400 °C (sample 15) display smooth surfaces with much smaller grains. The color bar shown on the left of the figure corresponds to measured sample height in nm. The scale bar shown on the bottom-right of each panel corresponds to a length of 200 nm.

tions, evident in Fig. 3(a). Grains are oriented either in parallel or $\pm 60^\circ$ relative to each other. Similar grain structures were observed in previous studies of high-temperature epitaxial grown Ta on c-plane sapphire substrates.^{15,25} In these studies, it was found that the grains aligned with the hexagonal (001)-oriented c-plane Al_2O_3 substrate and (110)-oriented Ta crystal. On the other hand, the control film grown on silicon with a Nb seed layer (sample 1) displays random grain orientations. Both control films have a low mean surface roughness of 0.518 nm and 0.72 nm, for films on silicon and sapphire, respectively.

Our different oxidation procedures resulted in oxide layers with varying surface morphologies and roughness. Thermal oxidation in the tube furnace at 400 °C resulted in the formation of an oxide layer composed of large grains – approximately 90-130 nm in length and 10-15 nm in width – that were randomly distributed in clustered formations. Within each cluster, the grains exhibited a nearly parallel alignment. The films treated through this process exhibited mean surface roughnesses (R_a) of 1.04 to 1.71 nm, indicating a consistently smooth surface morphology.

Films that underwent rapid thermal annealing at 700 °C had high mean surface roughnesses ranging from 7.28 to 16.38 nm. We note that within the $1 \times 1 \mu\text{m}^2$ region shown in Fig.

3 for a film that underwent 10 minutes of oxidation (sample 8), the mean surface roughness is 5.436 nm, compared to the higher value obtained from a $10 \times 10 \mu\text{m}^2$ scan ($R_a = 15.7$ nm). This indicates that the topography is non-uniform across lateral scales in the range of 1-10 μm , making this oxidation method unfavorable for the formation of reliable Josephson junction barriers, where a controllable and uniform oxide thickness is desired.

Plasma oxidation resulted in the smoothest films, with $R_a \approx 0.105 - 0.613$ nm and the lowest roughnesses observed for films oxidized at 200 °C. Based on the much lower surface roughnesses obtained by this method compared to thermal oxidation in the tube furnace, we determined that plasma oxidation is optimal for systematic growth of a smooth, high-quality oxide on tantalum for Josephson junction fabrication.

D. Ta Oxidation Mechanisms

To rationalize these results, we turn to ab initio modeling and compare the relative stability of partially oxidized Ta depending on the concentration and spatial distribution of the oxygen species. We considered three distinct scenarios for

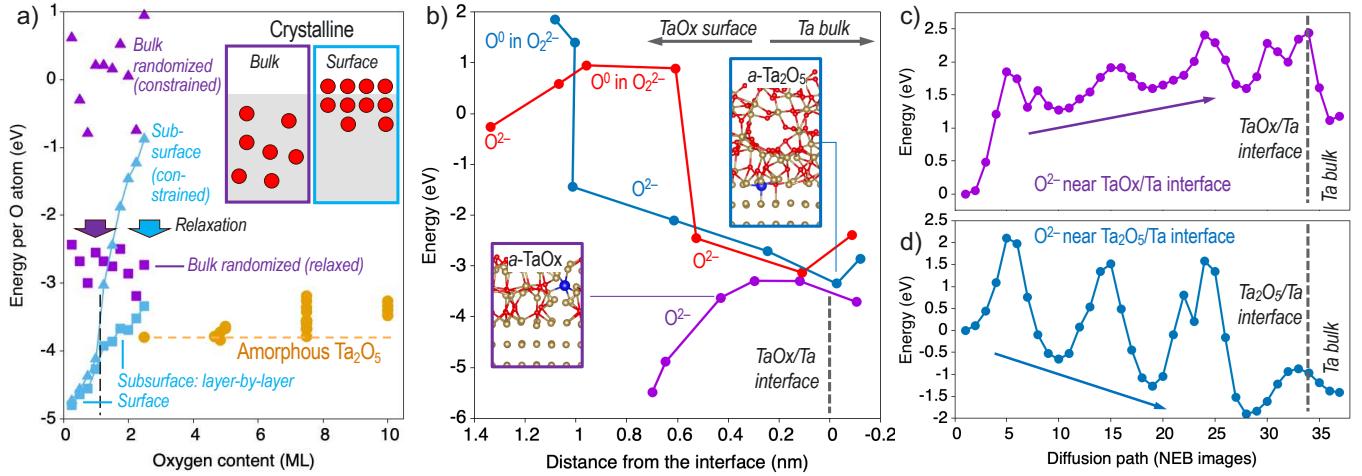


FIG. 4: (a) Thermodynamic stability of the TaO_x films supported on the Ta(110) surface depending on the content and spatial distribution of the oxygen species. Isolated O impurities in the bulk Ta are the least stable. Their presence induces lattice relaxation and promotes layer-by-layer accumulation of oxygen, which is further stabilized by the Ta oxidation to Ta₂O₅ and accompanying amorphization. (b) Energies of oxygen adsorption on the TaO_x surface and absorption into the TaO_x films, depending on the proximity to the interface and film composition. Oxygen adsorbs and diffuses as O²⁻ if the film is not fully oxidized or if the Ta interface is close enough to promote charge transfer, and as O⁰ bound to a preexisting O²⁻ and forming an O₂²⁻ molecular ion otherwise. (c) and (d) Potential energy profiles for the diffusion of interstitial O²⁻ in the case of partially oxidized TaO_x (c) and nearly fully oxidized amorphous Ta₂O₅ (*a*-Ta₂O₅) (d).

the oxygen species: (1) interstitial O injected into Ta disperse throughout the bulk; (2) oxygens sequentially occupy surface binding sites and subsurface interstitial sites at a 1:1 ratio with Ta, thus forming a TaO composition in the near-surface region; (3) oxygens are randomly distributed over the volume occupied by the surface Ta planes so as their ratio Ta:O=2:5. In the latter case, the lattice relaxations results in amorphous structures. Comparison of the calculated energy gain per O atom vs oxygen content for these scenarios [see Fig. 4(a)] shows that fully oxidized amorphous Ta₂O₅ is more stable than Ta with a spread-out oxygen distribution or quasi-ordered layer-by-layer TaO structures. Thus, isolated interstitial oxygen atoms are unlikely to diffuse from the surface into the Ta bulk. However, these atoms can be readily incorporated via oxygen plasma treatment, which would skew the native oxide formation.

For the plasma process, oxygenation of tantalum begins with the injection of oxygen ions, through the tantalum native oxide layer, penetrating to a depth defined by the stopping range — an extent that depends directly on the plasma power used during the process. Once implanted, these ions undergo thermal diffusion — our calculations suggest that the barrier for this diffusion is ~ 1 eV [see the last barrier in Fig. 4(d)] — and spread out over the Ta lattice. However, as their concentration increases, the strain buildup associated with interstitial atoms relaxes via the lattice expansion in the out-of-plane direction. This relaxation increases spacing between the Ta planes and promotes a layer-by-layer oxygen accumulation and eventual formation of amorphous Ta₂O₅ (*a*-Ta₂O₅) [see Fig. 4(a)].

We note that oxidation due to plasma treatment is accompanied by the oxygen intake from the surface. At low oxygen content, the adsorption and absorption of additional O species

is thermodynamically preferred (purple line in Fig. 4(b)) and their diffusion is facile with the calculated barriers of ~ 0.5 eV [Fig. 4(c)]. The uphill slope in Fig. 4(c) indicates the thermodynamic preference to saturate oxygen content locally, i.e., form Ta₂O₅, before diffusing toward the Ta metal.

As the Ta oxidizes to TaO_x, the oxygen deposition location changes due to the differences in the Ta and TaO_x stopping ranges and the film expansion; eventually, the newly injected oxygen ions are deposited within the oxide itself. If these oxygens are deposited in the vicinity of the interface between a nearly fully oxidized Ta₂O₅ and Ta metal, they draw the electron charge from the Ta substrate, convert to O²⁻ [Fig. 4(b)], and diffuse toward the interface driven by the positive charge associated with the interface. This trend is illustrated by the negative slope in (Fig. 4(d)). Importantly, the slope changes sign at the interface, indicating that further O²⁻ diffusion into metal Ta is suppressed⁴² even though the barrier for diffusion decreases from ~ 2 eV to ~ 1 eV.

As the lattice expansion proceeds and the local Ta:O ratio reaches 2:5, the new oxygen species are no longer deposited near the Ta interface. In this case, they form peroxy species (see Supplementary Information section S4), i.e., a neutral atom O⁰ binding to the lattice O²⁻ and forming an O₂²⁻ molecular ion. (For ball-and-stick representations of the local atomic configurations of excess oxygen in Ta₂O₅ and associated energy profile, see Supplementary Figure S4). At low concentrations, these neutral oxygen species do not have a preferential diffusion direction. Since their formation is unfavorable relative to the O₂ gas-phase molecule [blue and red lines in Fig. 4(b)], they are likely to escape the Ta₂O₅ oxide layer through out-diffusion.

Consequently, the final thickness of the oxide is governed by the plasma power, which sets the initial penetration depth and saturation time that depend on the kinetic energy of the plasma species and flux, and the temperature, which controls the extent of subsequent thermal diffusion. As the process continues, any additional oxygen may either remain interstitial or diffuse out of the film.

To compare the plasma and tube furnace oxygenation processes, note that the tube furnace operated at an oxygen flow rate nearly 1000 times higher and produced significantly thicker oxide layers. This elevated flow rate likely contributes to a high oxygen chemical potential, which promotes the formation of interstitial oxygen atoms and their diffusion deeper into the film owing to the high concentration gradients. For more details, refer to the discussion on the interstitial O^0 atom diffusion in α -Ta₂O₅ in Supplementary Information section S4.

E. Trilayers

This work was motivated by the need to develop growth procedures for insulating barriers used in Ta-based SIS Josephson junctions. Our final goal in this process is to grow α -Ta on top of this Ta₂O₅ layer, forming a SIS trilayer stack suitable for junction fabrication. A previous study demonstrated that a trilayer stack with a primarily α -phase top Ta layer can be achieved through heated deposition of Ta on top of an oxidized α -Ta base layer, but the junctions were suspected to have pinholes, resulting in micro-shorts.⁴⁴ Recognizing from our Ta film growth and characterization that α -Ta can be achieved either via nucleated growth using a Nb seed layer at room temperature or by direct deposition at 500 °C without a seed layer, we evaluated both approaches to determine their effectiveness in producing α -Ta on our oxide surfaces.

For one process, we sputtered a 6-nm Nb seed layer, followed by 100 nm of Ta on top of cuts of a Ta film with a native oxide (sample 1) and two Ta films that were oxidized in an oxygen plasma (samples 15 and 16). For the second process, we sputtered 150 nm of Ta on top of cuts of samples 1, 2, 15, and 16 held at 500 °C. The deposition rates, argon process pressure, and DC power are identical to those reported in the Methods section for the initial α -phase Ta deposition process.

Figure 5 shows the XRD spectra for these samples. Trilayers labeled as I and III were grown at room temperature with an Nb seed layer on samples 1 (control) and 16 (plasma oxidized), respectively. For the trilayers labeled IV and VI, the top Ta layer was deposited at 500 °C without a seed layer. Lastly, VII labels the trilayer in which Ta was also grown at 500 °C without a seed layer, but on sample 2 (control on sapphire).

Focusing first on the trilayers shown in panel (a), where tantalum growth on Ta₂O₅ was facilitated by an Nb seed layer (green curves labeled I and III), we observe distinct reflections corresponding to the (110), (211), and (220) planes of α -Ta. In the sample with a 14.88 nm oxide layer, a pronounced peak attributed to δ -Ta₂O₅ emerges at a 2θ angle of approximately 36.9°, while in the sample with a thinner 1.93 nm oxide, the magnitude of this peak is significantly reduced, manifesting only as a broadening of the adjacent (110) α -Ta peak. The

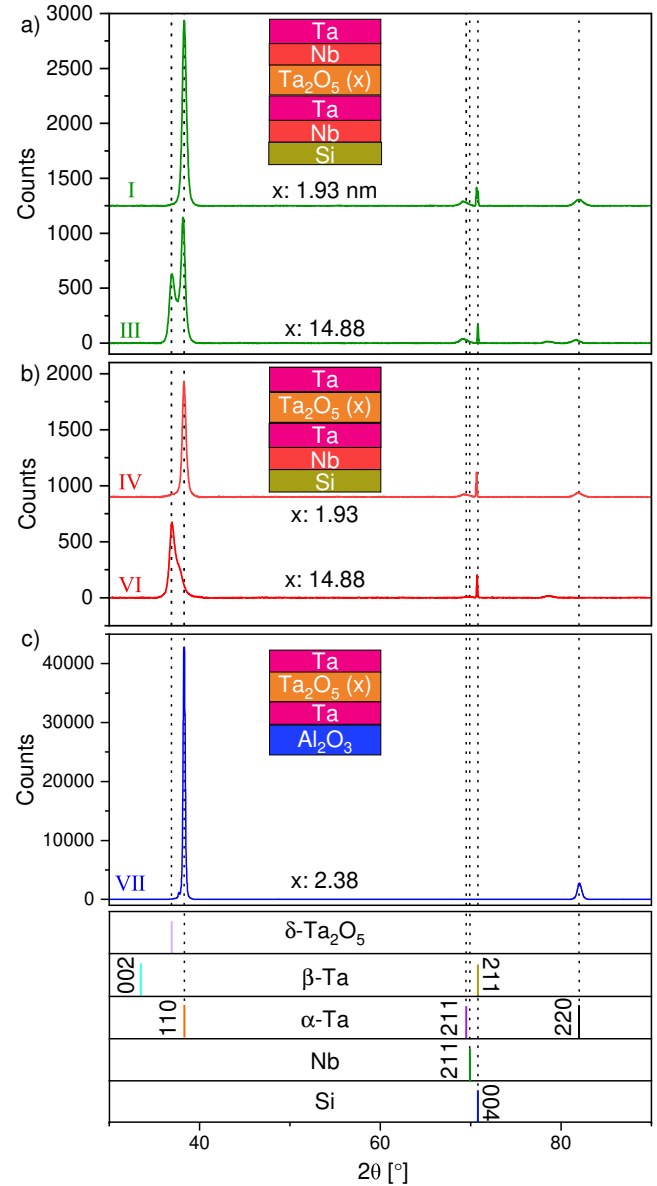


FIG. 5: (a-c) X-Ray diffraction (XRD) patterns of the trilayer films. The stack order for each set is shown in the cartoon insets, with "x" indicating the thickness of the oxide in nm. (Bottom panel) Reference peak angles for the phases of interest: δ -Ta₂O₅ (36.9°),⁴⁵ α -Ta (110, 38.3°),⁴⁶ (211, 69.1°),⁴⁷ and (220, 82°),⁴⁶ Nb (211, 69.1°),⁴⁸ and Si (004, 70.8°).²⁴ as measured in our films.

typical prominent (002) peak of β -Ta that appears in pure β -Ta⁴⁹ is notably absent, suggesting that we have no/negligent β -phase Ta in either layers. Regarding potential contributions from the Nb seed layer, it is important to note that the (211) reflection of Nb closely aligns with the (211) α -Ta peak, resulting in noticeable broadening around 69°-70° in the spectra. A substrate peak of Si (004) was observed at 70.8°, consistent with the (004) peak measured in our Si substrates [see Fig. S1(c)].

Figure 5(b) displays the X-ray spectra for films in which tantalum was deposited at 500 °C directly onto the Ta₂O₅ layer, without an Nb seed layer. While the same crystalline phases appear as in the previously discussed stack with Nb atop the oxide, the relative intensities of the peaks differ. Notably, the (110) α -Ta peak is significantly reduced, appearing as a broad shoulder for the δ -Ta₂O₅ peak because of the overlap between the two peak positions.

Lastly, Fig. 5(c) presents data for a trilayer in which the top tantalum layer was deposited directly onto Ta₂O₅ at 500 °C. Unlike the samples in panel (b), the bottom Ta layer in this structure was grown directly on a sapphire substrate, rather than on silicon with a niobium seed layer. The X-ray spectra reveal only the (110) and (220) reflections characteristic of α -Ta for this method.

In conclusion, we see that all three methods produce a top α -Ta layer, therefore can be used to grow trilayers for SIS junctions. To avoid a Nb interlayer, heated deposition processes are favored, if possible.

F. Transmission Electron Microscopy (TEM) of Trilayers

To evaluate the interface quality, we imaged three trilayer samples using scanning transmission electron microscopy, including high-angle annular dark-field (HAADF) imaging and energy-dispersive X-ray (EDX) spectroscopy, shown in Fig. 6. STEM imaging of sample VII (Ta/Ta₂O₅/Ta/Al₂O₃) revealed two Ta metal layers of 168 nm (top), and 160 nm (bottom) sandwiching a Ta₂O₅ layer with non-uniform thickness ranging from 4 – 8 nm and associated roughness at the interfaces. EDX elemental mapping in Fig. 6(d, g) shows small amounts of Ta-rich regions within the barrier, which may lead to superconducting shorts.

Figure 6(b) presents the STEM image for the only trilayer in which the top layer was grown at room temperature – sample I, which consists of the stack Ta/Nb/Ta₂O₅/Ta/Nb/Si. STEM imaging and EDX mapping [Fig. 6(e,h)] imaging showed a thin layer of Ta₂O₅ of thickness between $\sim 2 - 4$ nm between two $\sim 100 - 105$ nm thick Ta layers. The top Nb layer used to nucleate an α -phase top electrode measured at 13 nm thick, whereas the bottom Nb seed layer atop the substrate appeared to be 5-6 nm.

Lastly, Figs. 6(c, f, i) display the STEM and EDX images of sample V, which contains the Ta/Ta₂O₅/Ta/Nb/Si stack. The Ta₂O₅ layer appears uniform, 15 nm thick, and has sharp interfaces. For all samples, EDX analysis detected oxygen signals throughout the top and bottom Ta layers (see Supplementary Fig. S3). This contamination of oxygen in the Ta bulk likely occurred due to exposure of the FIB-cut lamella to air prior to STEM imaging and EDX mapping.

IV. CONCLUSIONS

Through thermal oxidation in a tube furnace, rapid thermal annealing, and plasma oxidation, we grew Ta₂O₅ on top of sputtered α -Ta thin films. In particular, plasma oxidation created

smooth oxide surfaces conducive to the deposition of a second Ta layer on top to form trilayer structures. The mechanistic insights into the Ta oxidation process provide guidance for the modulation of the Ta oxide composition and thickness by on-the-fly tuning the plasma parameters. We also demonstrated successful growth of α -Ta on top of both native and plasma-oxidized α -Ta films, paving the way for trilayer Josephson junctions based on Ta and Ta₂O₅.

Our results suggest that we can perform in-situ growth of Ta/Ta₂O₅/Ta trilayer structures without breaking vacuum. These structures may be used to fabricate highly oriented Ta-based Josephson junctions, which could significantly lower the density of TLS defects in the barrier and at interfaces.^{50,51} Superconducting qubits incorporating such junctions may demonstrate improved lifetimes and coherence beyond current state-of-the-art Ta-based qubits that utilize conventional Al-based junctions.

V. METHODS

A. DC Magnetron Sputtering of Tantalum Films

We employed two distinct methods to deposit α -phase tantalum films. For both procedures, the chamber base pressure was reached below 6×10^{-8} Torr prior to heating. All substrates were pre-cleaned using acetone and isopropanol, then dried with a nitrogen (N₂) spray.

In the first approach, we sputtered tantalum onto 430 μ m-thick, double-side polished sapphire substrates first heated to 500 °C at a rate of 20 °C per minute, and thermally degassed at 500 °C for 2 hours. Following degassing, we deposited a 150 nm Ta layer while maintaining the substrate temperature at 500 °C. The substrates were then cooled to room temperature at a rate of approximately 12 °C per minute.

The second method involved sputtering tantalum at room temperature onto Si(100) substrates with thicknesses of either 500 μ m or 655–695 μ m. We first deposited a 6 nm niobium seed layer, followed by either 60 nm or 100 nm of tantalum. The DC sputtering powers were set to 510 W for Nb and 310 W for Ta. Both materials were deposited under an argon process pressure of 3 mTorr, yielding a deposition rate of approximately 15 nm per minute.

B. X-ray Spectroscopy, Diffraction, and Reflectometry

We acquired XPS spectra using a Kratos Axis-Ultra DLD spectrometer equipped with a monochromatized Cu K α X-ray source and a low-energy electron flood gun for charge neutralization. During spectral acquisition, the analytical chamber pressure remained below 5×10^{-9} torr. Survey and compositional spectra were recorded at a pass energy of 80 eV, while high-resolution spectra were collected at a pass energy of 20 eV. The take-off angle—defined as the angle between the sample normal and the energy analyzer’s input axis—was set to 0°, corresponding to an approximate sampling depth of 100 Å. The Kratos Vision2 software program was used to determine

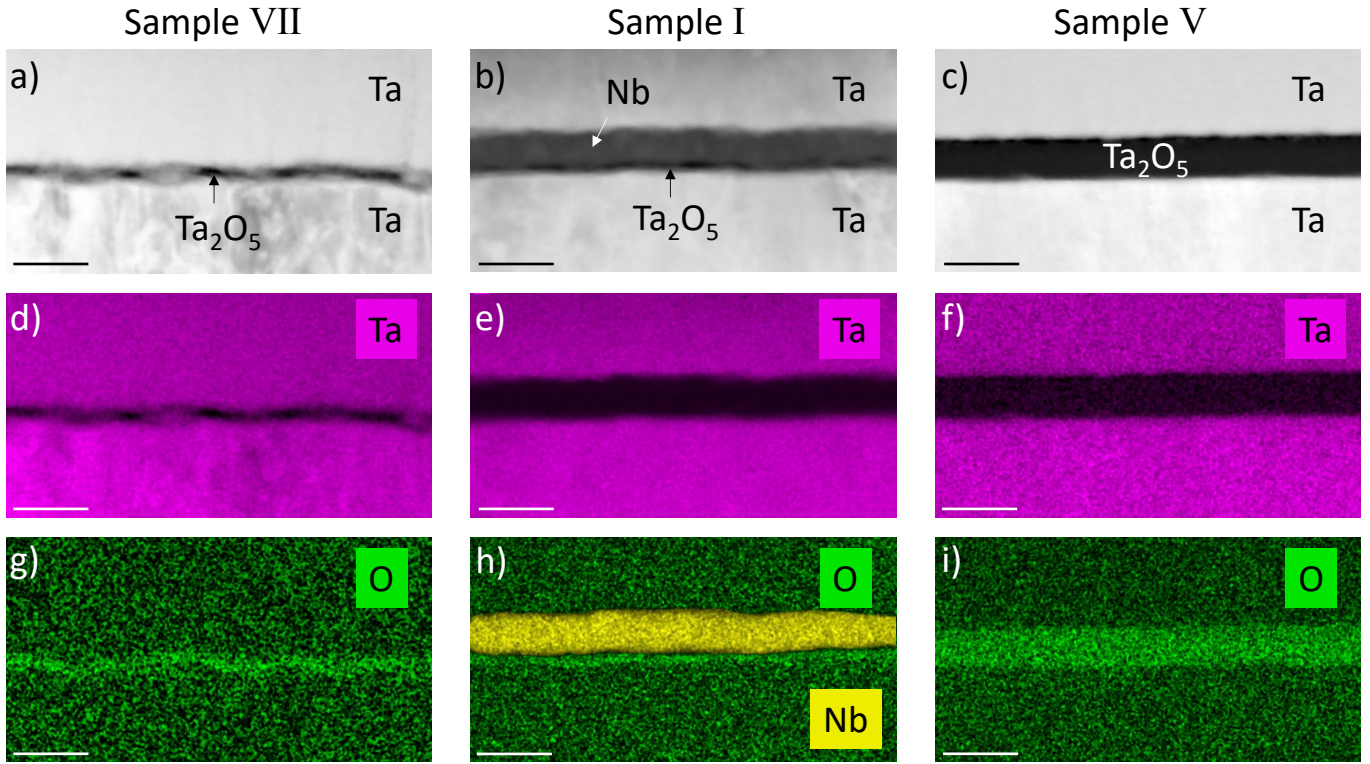


FIG. 6: [Top row] STEM-HAADF overview images of three different trilayer samples, corresponding to (a) sample VII and (b) I, as labeled in Fig. 5, as well as (c) sample V (XRD absent from Fig. 5). [Middle row, d - f] STEM-EDX elemental mapping of Ta for the three samples. [Bottom row, g - i] STEM-EDX elemental mapping of O for the samples (and Nb for sample I).

peak areas and to calculate the elemental compositions from peak areas. CasaXPS was used to peak fit the high-resolution spectra. For the high-resolution spectra, a Shirley background was used, and all binding energies were referenced to the C 1s C-C bonds at 285.0 eV.

XRD analysis was done using Bruker D8 Discover with a Cu anode X-ray source (50kV, 1000 μ A), combined with a Pilatus 100K 2D detector. The experimental XRD data was then processed using DIFFRAC.EVA software package.

For XRR analysis, the same instrument with the same X-ray source, equipped with a Lynxeye-XET multi-strip 1D detector was used, with a 0.2 mm beam collimator. The experimental XRR data were fitted to a theoretical model using the DIFFRAC.XRR software package. The layer thicknesses, densities, and interface roughnesses were varied to achieve the best fit to the experimental data. The goodness of fit was determined by minimizing the χ^2 value between the measured and simulated reflectivity curves.

C. Atomic Force Microscopy (AFM)

Atomic force microscopy was performed using a Bruker ICON AFM, with either a Bruker ScanAsyst-Air probe with a 2 nm tip in ScanAsyst mode, or a Bruker OTESPA-R3 probe with a 7 nm tip in tapping mode. The 1 μm^2 scans were collected at a rate of 1 Hz with 512 samples per line, and the

10 μm^2 scans were collected at a rate of 1 Hz with either 256 or 512 samples per line.

D. Electron Microscopy

TEM samples were prepared using an FEI Helios NanoLab 600i Focused Ion Beam Scanning Electron Microscope (FIB/SEM). Layer stacking and elemental composition were examined with a Talos F200X STEM operated at 200 kV, equipped with a field emission gun (FEG) and four in-column Super-X EDS detectors. STEM-HAADF (high angle annular dark-field) images were recorded using a convergence semi angle of 10.5 mrad.

E. Computational Modeling

The dependence of the thermodynamic stability of Ta oxide films supported on the Ta substrate on the content and spatial distribution of the oxygen species was quantified using *ab initio* simulations within the density functional theory (DFT), and a generalized gradient approximation exchange correlation functional⁵² as implemented in the VASP code.⁵³ The plane wave augmented pseudopotentials were used.⁵⁴ The Ta slab was terminated with (110) surfaces and included 8 atomic planes; the lateral supercell (9.33 $\times$ 9.90 $\text{\AA}^2$) included 12 Ta

atoms in each plane. The out-of-plane lattice parameter was fixed at 42 Å, which leaves at least 15 Å wide vacuum gap even after accounting for the expansion due to Ta oxidation. The structural models of the amorphous Ta₂O₅ (*a*-Ta₂O₅) and Ta oxide (TaO_x) film on the Ta surface were generated by incorporating oxygen atoms into the Ta slab.

Since Ta₂O₅ and TaO_x are disordered, the simulations were conducted at the Γ point only; the plane wave basis set was fixed at 500 eV. The total energies were minimized with respect to the internal atomic coordinates, except for the Ta atoms of the furthers from the interface plane, which remained fixed at their crystallographic sites. The total energy conversion criterion was set to 10⁻⁵ eV. Energies of oxygen interaction with Ta and Ta oxides are calculated relative to the energy of the gas-phase O₂ molecule. The energy barriers for oxygen diffusion were calculated using the nudged elastic band (NEB) method.⁵⁵ The atomic charges were analyzed using the method developed by Bader.⁵⁶

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AUTHOR CONTRIBUTIONS

S.E. and D.P.P. conceived and designed the experiment. R.P. and R.T. grew the tantalum films and performed the oxidations, X-ray reflectometry, atomic force microscopy, scanning electron microscopy, and magnetometry measurements. S.B. col-

lected some X-ray reflectometry data and provided advice on extracting accurate oxide-layer thicknesses. TEM specimens (lamellae) were prepared using the FIB lift-out in cross-section by A.B. P. K. performed the STEM-EDX measurements and carried out the data interpretation of these samples. R.P. performed data analysis. P.V.S. determined the oxygenation pathways and oxidation mechanisms of the Ta films. S.E., R.P., R.T., and P.V.S. wrote the manuscript. All authors commented on the manuscript.

COMPETING INTERESTS

The authors have no conflicts to disclose.

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Supplementary Information: Fabrication and Structural Analysis of Trilayers for Tantalum Josephson Junctions with Ta₂O₅ Barriers

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S1. Magnetization Measurements and X-Ray Diffractometry of Control Samples

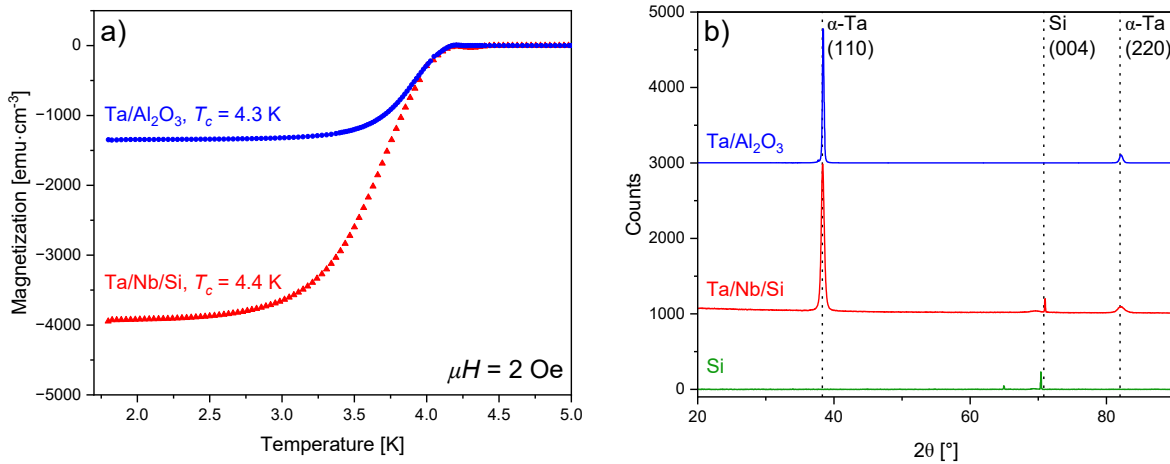


Figure S1: **a)** Temperature-dependent DC magnetization measurements of both (Si, Al₂O₃) control Ta films, showing superconducting transitions. **b)** XRD spectra of control Ta films grown on on Al₂O₃ and Si substrates, as well as a bare Si substrate.

Magnetization measurements were performed to determine the critical temperature T_c of the tantalum films. Figure S1 shows the magnetization data of the two control samples: 100-nm Ta film sputtered at room temperature on top of a 6-nm seed layer on a Si substrate and 150-nm Ta film sputtered on a sapphire substrate held at 500 °C. Both samples exhibit diamagnetic transitions, characteristic of the Meissner effect, with critical temperatures of 4.4 K and 4.3 K, respectively. The X-ray diffraction (XRD) spectra of the control Ta films grown on both sapphire (Al₂O₃) and silicon (Si) substrates demonstrate narrow peaks suggestive of high film quality, with prominent α -Ta (110) and (220) peaks at 38.3° and 82°, respectively. To identify the XRD peak associated with the Si substrate, we also performed XRD on a bare Si wafer. We observe a clear Si (004) peak at 70.8° that is also present for the Ta film grown on Si.

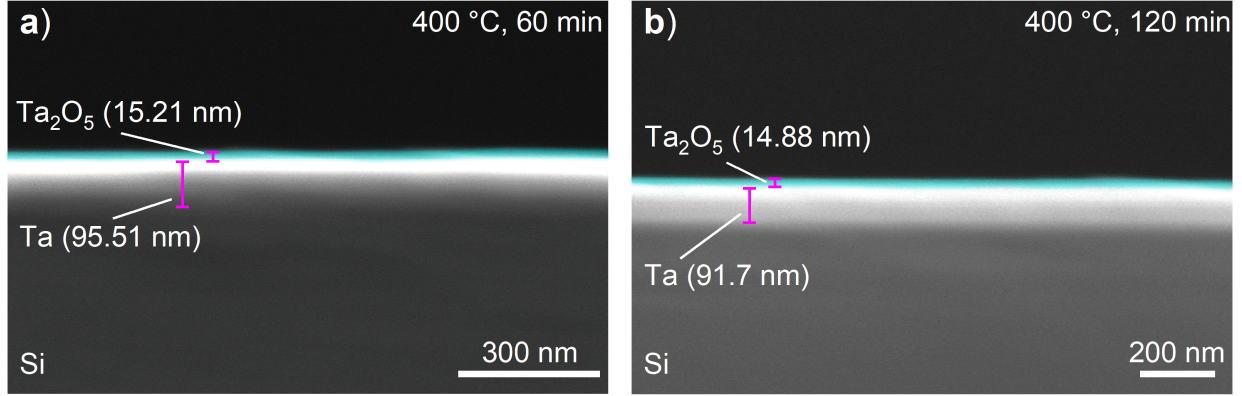


Figure S2: SEM images showing cross sections of tantalum films that underwent plasma oxidation at 400 °C for **a)** 60 min and **b)** 120 min. The Ta₂O₅ layer is highlighted in false color, and the thickness values of the Ta and Ta₂O₅ layers extracted from XRR measurements are labeled. The tantalum film on both samples was initially deposited at room temperature on a 6-nm Nb seed layer, which is more difficult to distinguish from the Si substrate due to insufficient contrast.

S2. Scanning Electron Micrographs of Plasma-Oxidized Ta Films

Cross sections of plasma-oxidized Ta films imaged through scanning electron microscopy (SEM) are shown in Fig. S2. The observed thicknesses of the Ta and Ta₂O₅ layers in SEM correspond well to the thickness values extracted from X-ray reflectometry (XRR) measurements. Images were collected using a ThermoFisher Apreo SEM at a working distance of 3 mm.

S3. Scanning Transmission Electron Microscopy

Energy-Dispersive X-ray (EDX) spectroscopy shown in Fig. S3 quantifies oxygen distribution across the Ta/Ta₂O₅/Ta stack. Spectra were acquired from three regions: top Ta electrode, Ta₂O₅ barrier, and bottom Ta electrode. The oxygen peak at 0.525 keV shows maximum intensity (~ 2500 counts) in the Ta₂O₅ barrier as expected. However, both Ta electrodes exhibit substantial oxygen peaks of ~ 1900 counts (top Ta) and ~ 2100 counts (bottom Ta)—only slightly lower than the barrier itself. This indicates oxygen is not confined to the intentional oxide layer but is present throughout both metallic electrodes. The oxygen likely exists as dispersed oxide phases, interstitial atoms, or grain boundary segregation within the Ta layers.

S4. Interstitial Oxygen Diffusion in α -Ta₂O₅

Excess oxygen in fully oxidized amorphous Ta₂O₅ takes the form of a peroxy species (O_2^{2-}), i.e., a neutral oxygen O^0 atom bound to a lattice O^{2-} ion in a dumbbell configuration. To determine the oxygen diffusion barrier, we first generated a model α -Ta₂O₅ structure, represented by a Ta₃₆O₉₀ supercell. The initial structure was obtained by randomly distributing Ta and O atoms over the supercell volume. Then the total energy of this structure was minimized with respect to the atomic coordinates and supercell parameters; the resulting structure was further modified by applying atomic displacements and followed by one more energy minimization step. The displacement directions were selected at random, and the magnitude of the displacements was varied (also at random) between 1 and 3 Å. The lowest energy configuration was selected for further calculations.

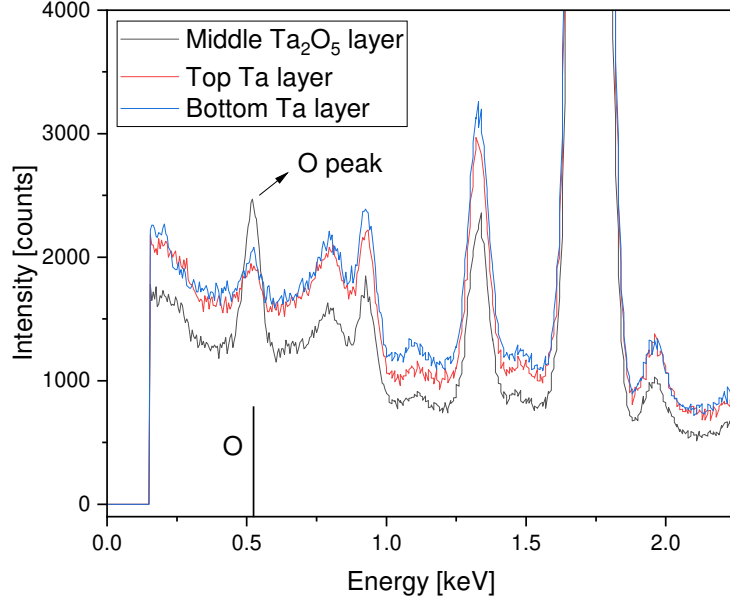


Figure S3: EDX spectra (of Ta/Ta₂O₅/Ta stack, Sample VII from Fig. 4) from top Ta electrode (red), Ta₂O₅ barrier (black), and bottom Ta electrode (blue). Oxygen peak (0.525 keV) present in all three layers, confirming bulk contamination in Ta electrodes beyond the intentional barrier region.

The stability of point defects in an amorphous lattice depends on the details of their local environment. We analyzed the effect of this variability on the stability of excess O⁰ by considering four configurations and calculating six diffusion barriers between these configurations (see Fig. S4). Here, the O species participating in the diffusion process are shown with large colored spheres (purple, blue, cyan, green, and orange). The initial configuration (A) is formed by the purple-blue dumbbell O₂²⁻ ion. Transition to configuration B corresponds to breaking the purple-blue bond and forming the blue-cyan bond, and so on, as shown in panels C (cyan-green) and D (green-orange). Our sampling indicates that the O diffusion barrier varies between 0.6 and 1.2 eV, depending on the local configuration, which points to the possibility of a facile O diffusion at elevated temperatures. These calculations also suggest that the formation of interstitial oxygen is thermodynamically unfavorable relative to the gas-phase O₂ molecule by 0.8–1.4 eV. Thus, we conclude that forming an appreciable concentration of these ions is possible only under high oxygen chemical potential conditions, such as high O₂ pressure and elevated temperature. However, once formed and in the presence of high concentration gradients, these species provide a potent channel for enhanced Ta oxidation that is not available at normal conditions.

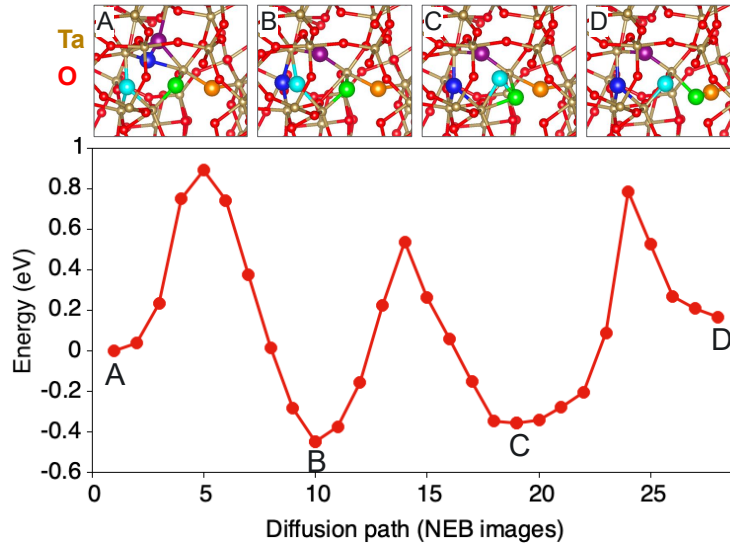


Figure S4: Ball-and-stick representations of the local atomic configurations of the excess oxygen atom along its diffusion path in $a\text{-Ta}_2\text{O}_5$ (top) and the corresponding potential energy profile (bottom). Large colored spheres indicate five oxygen atoms participating in the considered interstitialcy oxygen diffusion. In all cases, the excess O^0 atom binds to a lattice O^{2-} , thus forming a dumbbell O_2^{2-} configuration. The diffusion of O^0 proceeds by breaking one dumbbell configuration, displacing the atom toward a neighboring O^{2-} and forming a new dumbbell configuration, as indicated with the rearrangement of the large colored spheres along the A–B–C–D path.