

Structure and Formation Mechanisms in Tantalum and Niobium Oxides in Superconducting Quantum Circuits

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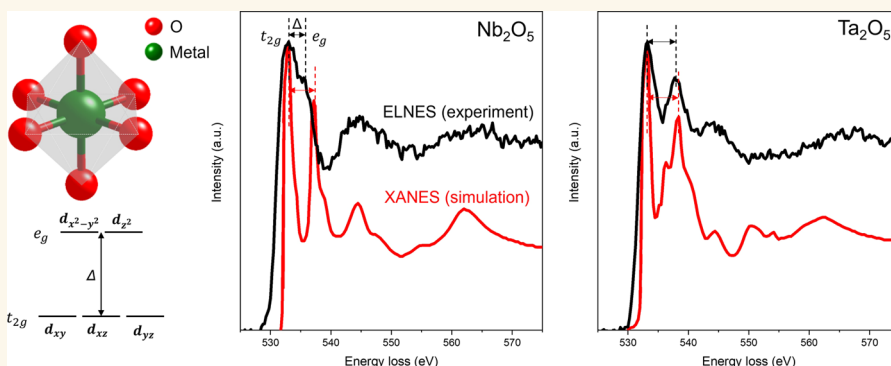
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ABSTRACT: Improving the qubit's lifetime (T_1) is crucial for fault-tolerant quantum computing. Recent advancements have shown that replacing niobium (Nb) with tantalum (Ta) as the base metal significantly increases T_1 , likely due to a less lossy native surface oxide. However, understanding the formation mechanism and nature of both surface oxides is still limited. Using aberration-corrected transmission electron microscopy and electron energy loss spectroscopy, we found that Ta surface oxide has fewer suboxides than Nb oxide. We observed an abrupt oxidation state transition from Ta_2O_5 to Ta, as opposed to the gradual shift from Nb_2O_5 , NbO_2 , and NbO to Nb, consistent with thermodynamic modeling. Additionally, amorphous Ta_2O_5 exhibits a closer-to-crystalline bonding nature than Nb_2O_5 , potentially hindering H atomic diffusion toward the oxide/metal interface. Finally, we propose a loss mechanism arising from the transition between two states within the distorted octahedron in an amorphous structure, potentially causing two-level system loss. Our findings offer a deeper understanding of the differences between native amorphous Ta and Nb oxides, providing valuable insights for advancing superconducting qubits through surface oxide engineering.

KEYWORDS: superconducting qubits, surface encapsulation, tantalum oxides, niobium oxides, STEM-EELS, CALPHAD

Quantum information processing holds the potential to surpass traditional computers in various aspects,^{1,2} including quantum physics, chemistry simulations,^{3,4} and optimization algorithms.⁵ Among the diverse materials considered for quantum computing, superconducting materials offer significant possibilities for solid-state platforms.^{6,7} Their fabrication utilizes well-established semiconductor methodologies,⁸ such as thin film deposition and lithography, which place a superconducting quantum bit (qubit) at the forefront of quantum computing technology.⁹

Over the last two decades, the advancement of superconducting qubits has primarily stemmed from optimizing the design of the quantum circuit.^{10–14} However, it is widely

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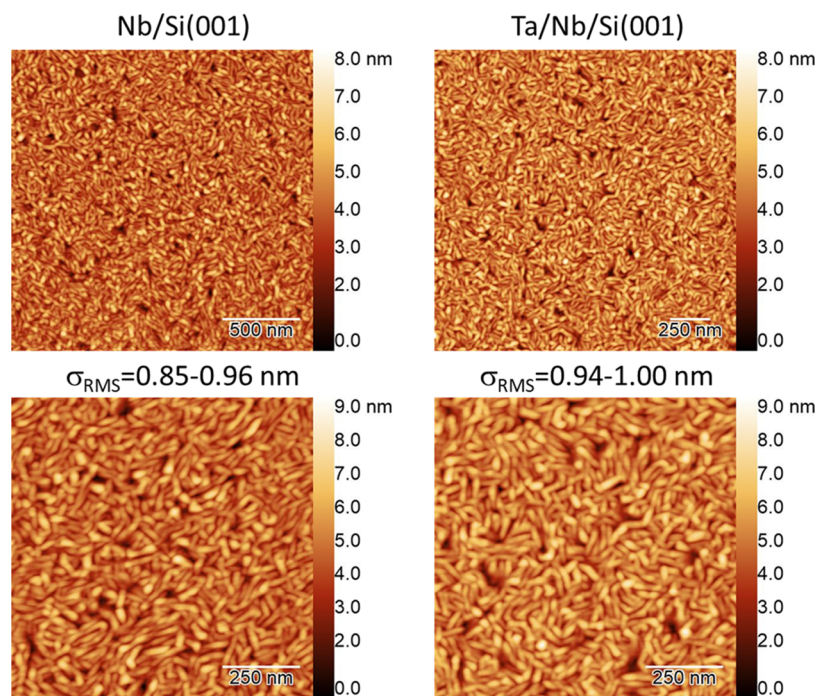


Figure 1. AFM surface images of Nb/Si and Ta/Nb/Si samples.

acknowledged that intricate processing methodologies can introduce defects at the interfaces and surfaces of the superconducting quantum circuits, thereby introducing decoherence sources.^{15–18} Therefore, a precise understanding of how the material's structure and defects contribute to decoherence is essential for enhancing the performance of superconducting qubits. This understanding is particularly crucial for the Josephson junction (JJ) and resonators, which serve as fundamental components in the superconducting quantum circuits.¹⁹

Niobium (Nb)-based superconducting quantum devices have dominated the field for an extended period as this material offers cost advantages, flexibility in deposition methodologies, and compatibility with industrial-scale processes.^{20,21} Nonetheless, as the T_1 of two-dimensional transmon qubits composed of Nb has long plateaued around 100 μs , it has become clear that alternative materials approaches are needed.²² Recent work highlights Nb surface oxides' critical role in limiting T_1 .²³ As a result, encapsulating the surface of niobium with various metals and dielectrics allows for significant improvements, with 2–5 \times longer T_1 achievable compared to bare Nb. Through this approach, Bal et al. achieved a maximum T_1 of 602 μs , with a tantalum (Ta) capping layer on Nb.²³ This demonstrates that Ta oxide is less lossy than Nb oxide and explains why other groups have previously been able to achieve max T_1 values of 360 μs (Place et al.,²⁴ reported Ta-based qubits) and 503 μs (Wang et al.,²⁵ demonstrated a further improvement through the implementation of a dry-etch process) with Ta. This improvement has been attributed to factors such as a thinner total surface oxide thickness,²⁶ a reduced number or absence of stable suboxides on the Ta surface,^{21,25–27} or the abrupt transition from higher oxidation states to lower ones.^{21,26} However, Bal et al.²³ suggest no clear correlation between oxide thickness and overall loss. Instead, they propose that this loss may arise from variations in the chemical stoichiometry (i.e., the presence of stable suboxides in the Nb oxide). Other potential factors

include variations in the local crystallinity and the presence of impurities. Despite these extensive studies, the reasons for forming fewer suboxides in Ta and their lower dissipative nature in terms of dielectric loss remain unclear. Furthermore, as niobium pentoxide is the most detrimental part of qubit's coherence,^{17,28,29} the low-loss characteristics of tantalum pentoxide may be another reason for performance enhancement. Thus, a direct comparison between the Ta and Nb pentoxides should be carried out thoroughly.

We systematically compare microstructure and device performance fabricated from two superconducting thin films, as discussed by Bal et al.:²³ (1) an uncapped Nb thin film on a silicon (Si) substrate (later called the uncapped film) and (2) Ta-capped Nb thin film on Si. Our results suggest that Ta oxide is less lossy than Nb oxide due not only to the formation of less complicated oxides but also to a closer-to-crystalline nature of the oxide bonding. Our results offer valuable insights for advancing superconducting qubits through surface oxide engineering.

RESULTS AND DISCUSSION

As discussed, Ta capping of Nb leads to a 2–5 \times improvement in the qubit T_1 .²³ This enhanced T_1 suggests that the notable systematic improvements are intricately linked to the metal-air (M-A) interface rather than the superconducting metal itself.²³ To evaluate the origin of the differences in loss between Ta and Nb oxides, we performed a detailed comparison of the M-A interface for both the Nb and Ta-capped Nb films.

Overall, the surface morphologies are nearly identical between the two samples. Figure 1 shows the atomic force microscopy (AFM) images of the Nb- and Ta-capped samples. The film's root-mean-square (RMS) roughness is less than 1 nm, exhibiting a similar value between the bare Nb and Ta-capped Nb thin films.

Cross-sectional transmission electron microscopy (TEM) micrographs in Figure 2 show that both samples exhibit

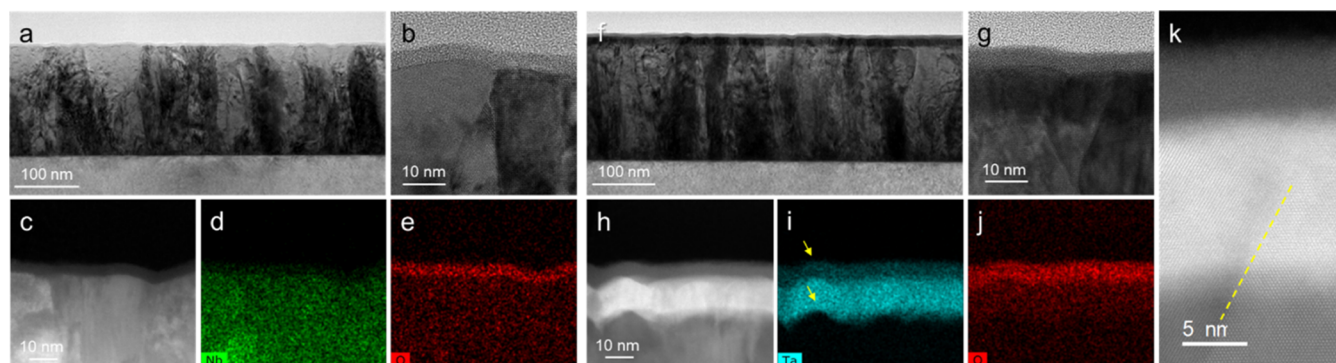


Figure 2. Cross-sectional TEM images of bare Nb and Ta-capped Nb thin films. a, Cross-sectional bright-field TEM image and b, high-resolution TEM image of the bare Nb thin film on Si. c, HAADF-STEM image and d, e, the corresponding EDS elemental distribution maps of Nb and O at the NbO_x-Nb interface. f, Cross-sectional bright-field TEM image and g, high-resolution TEM image of the Ta-capped Nb thin film on Si. h, HAADF-STEM image and i, j, the corresponding EDS elemental distribution map of Ta and O at the TaO_x-Ta-Nb interface. k, Atomic-resolution HAADF-STEM showing epitaxial growth of Ta on Nb.

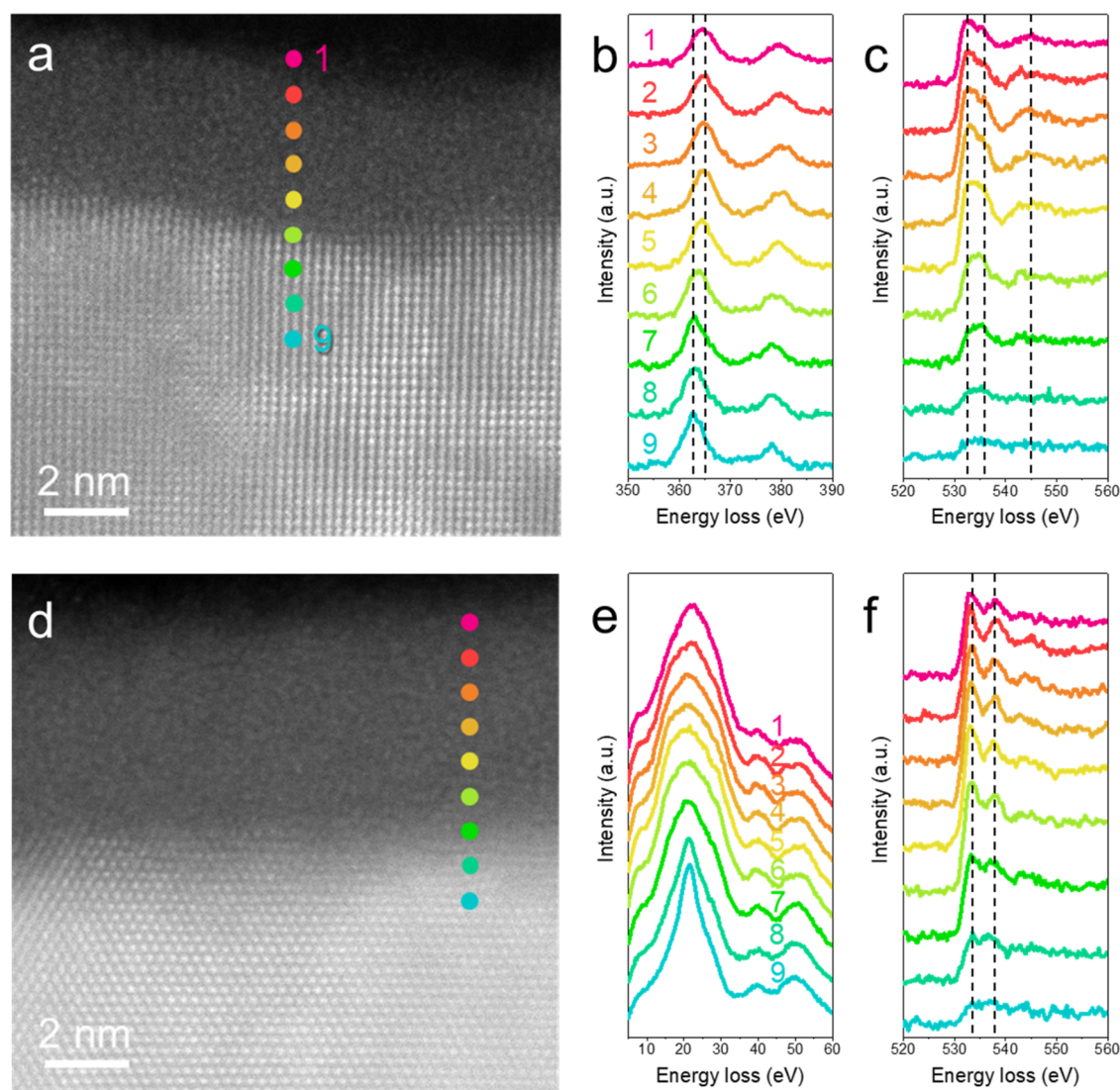


Figure 3. EELS line profiles of surface oxides on Nb and Ta. a, HAADF-STEM micrograph of the NbO_x-Nb interface of the bare Nb thin film. Colored spots denote the positions in which EEL spectra are taken. b, c, Corresponding spatially resolved EEL spectra showing ELNESs of Nb M_{2,3} edge and O K edge. d, HAADF-STEM micrograph of the TaO_x-Ta interface of Ta-capped Nb thin film. Colored spots denote the positions in which EEL spectra are acquired. e, f, Corresponding spatially resolved EEL spectra showing ELNESs of Ta O_{2,3} edge and O K edge. The distance between adjacent spots is 0.83 nm.

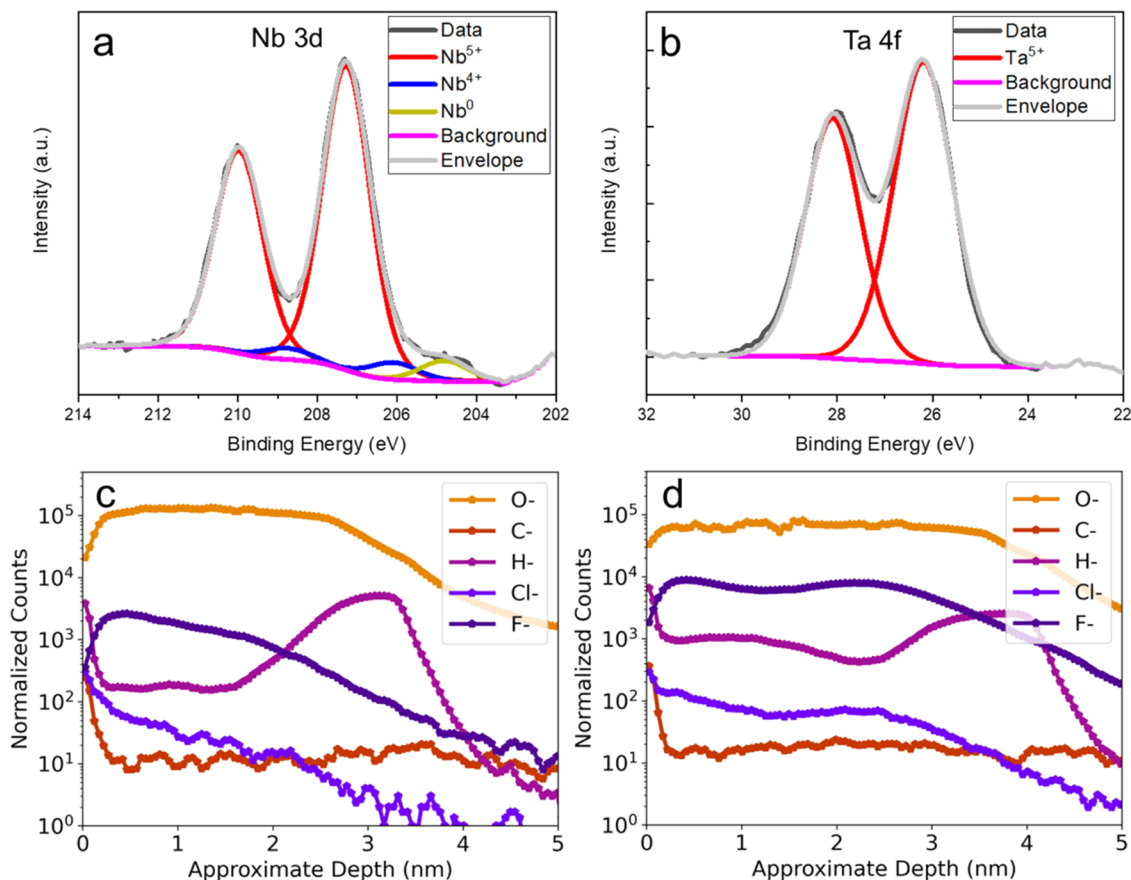


Figure 4. XPS and ToF-SIMS characterization. **a**, Nb 3d intensity consists of Nb₂O₅, NbO₂, and Nb (only Nb(0) 3d 3/2 is shown). **b**, Ta 4f intensity only consists of Ta₂O₅. Black, violet, and gray lines represent the raw spectrum, background, and envelope, respectively. Normalized ToF-SIMS depth profiles showing O⁻, C⁻, H⁻, Cl⁻, and F⁻ of **c**, bare Nb, and **d**, Ta-capped Nb thin films.

columnar grain structures, with the Nb thin films measuring approximately 200 nm thick and the Ta capping layers at around 10 nm. Notably, there is a thickness difference in the amorphous surface oxide layer between the two samples. The native amorphous surface oxide layer on the uncapped Nb film is approximately 4 nm. In comparison, the surface oxide layer on the Ta capping layer is approximately 5.5 nm (Figure 2b,g). In addition, the Ta-capped film's surface appears to follow the uncapped film's surface morphology, which aligns with the AFM analysis in Figure 1. Energy-dispersive X-ray spectroscopy (EDS) elemental distribution maps highlight these features, as depicted in Figure 2c–e,h–j. Atomic-resolution high-angle annular dark-field (HAADF)-scanning TEM (STEM) micrograph confirms the epitaxial growth of Ta on Nb due to its almost identical lattice constant, as shown in Figure 2k. The grain boundary of the Ta capping layer follows that of the Nb film, as indicated by a yellow dashed line in Figure 2k, which indicates that the Ta capping layer exhibits the same lateral grain size as the Nb thin film.

We further compared the oxygen states of Nb and Ta across the M-A interface. Figure 3 shows spatially resolved electron energy loss spectroscopy (EELS) results to elucidate the chemistry of the amorphous oxide layer. A gradual oxidation state transition is observed across the M-A interface for the amorphous Nb oxide layer. Cross-sectional HAADF-STEM images at the M-A interface indicate the spot position for spatially resolved EELS, where the distance between adjacent spots is 0.83 nm. Figure 3b displays the energy-loss near-edge

structure (ELNES) of the Nb M₂₃ edge, confirming a transition from +5 to +0, indicated by two dashed lines at 365 and 363 eV, respectively. According to the Nb M₂₃ edge,³⁰ the outermost four spots (1–4) correspond to the Nb₂O₅, followed by NbO₂ at spot 5. This gradual transition in the oxidation state is further supported by ELNES of the O K edge (Figure 3c), where the first two dominant peaks (indicated by dashed lines at 532.5 and 535.5 eV) with a third peak at 545 eV represent higher oxidation states (+4 and +5).³⁰ The intensity ratio of the first two peaks changes slightly at spot 5, indicating a decrease in oxidation state to +4, i.e., NbO₂.³⁰ This is consistent with our previous study³¹ and reveals a gradual transition in the amorphous oxide layer from Nb₂O₅ to NbO₂ and finally NbO (spot 6) at the interface between the amorphous surface oxide layer and Nb metal.

In contrast, the Ta surface exhibits a relatively abrupt transition from Ta₂O₅ to Ta. Unlike Nb, Ta lacks a core-loss edges fingerprint to track the oxidation states. However, the plasmon peaks in the low-loss energy region show an abrupt transition from spot 7 to spot 8 (Figure 3e), where spots 8 and 9 exhibit sharp plasmon peaks corresponding to metallic Ta, and spots 1 to 7 show bell-shaped plasmon peaks. The O K edge further confirms this abrupt transition, with two well-separated dominant peaks at 533.5 and 537 eV, denoted by dashed lines in Figure 3f, indicating Ta₂O₅ for spots 1 to 7.^{32,33} Therefore, based on the ELNES study across the Ta oxide layer, spot 8 may correspond to either suboxide species or a mixture of metallic Ta and some Ta₂O₅. To corroborate this,

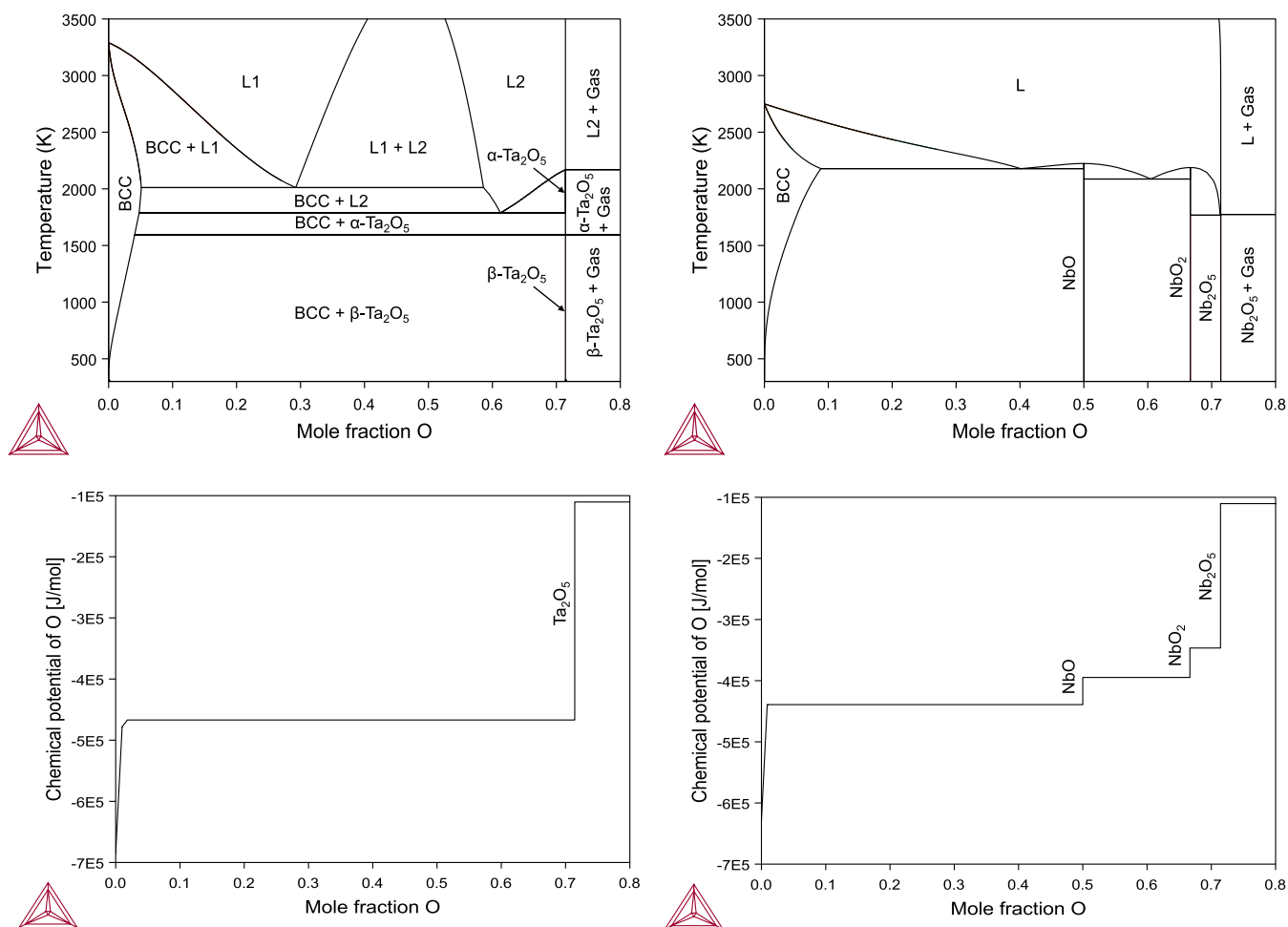


Figure 5. Calculated phase diagrams of the Ta–O and Nb–O systems and their respective oxygen chemical potentials as functions of oxygen mole fraction.

we performed principal component analysis (PCA). However, the PCA result did not show clear evidence of suboxide species at the Ta-TaO_x interface (details in the [Supporting Information](#)). In general, it is evident that Ta exhibits a thinner transition layer with an upper limit of 0.8 nm and undergoes a more abrupt transition from +5 to +0 states compared to Nb (within about 0.8–1.6 nm). This observation holds true, whether conservatively or radically determined, despite the total thickness of the amorphous Ta surface oxide layer being greater than its Nb counterpart, aligning well with previous works.^{23,25,26}

The X-ray photoelectron spectroscopy (XPS) analysis is in good agreement with the EELS results. [Figure 4a](#) shows NbO₂ and metallic Nb, although most of the Nb 3d intensity is Nb₂O₅. We do not observe a signal attributable to the NbO peak, maybe due to the small volume fraction. On the other hand, [Figure 4b](#) shows that Ta 4f is entirely explained by Ta₂O₅ peak fits. The difference in oxide thickness may be the main reason why there are no peaks from metallic Ta, as well as Ta having a much higher atomic number that results in a shorter mean free path of electrons. It might imply that even if there are some suboxides, photoelectrons cannot escape to be detected due to the thicker Ta oxide layer. Nonetheless, with the same X-ray interaction conditions for the Nb and Ta surface oxides, the Ta-capped sample shows uniform surface oxide in the form of Ta₂O₅. In contrast, the baseline Nb sample

exhibits a suboxide layer between predominant Nb₂O₅ and metallic Nb thin film, consistent with spatially resolved ELNES results in [Figure 3](#).

To examine the impurity content in the two surface oxides, we used time-of-flight secondary ion mass spectrometry (ToF-SIMS) to capture the signal associated with species such as C⁻, H⁻, F⁻, and Cl⁻ as a function of depth for both the uncapped and Ta-capped Nb films, as shown in [Figure 4c,d](#). Regarding H⁻ counts, we observe a clear increase in the signal close to the oxide and metal interface for both samples. As previously reported,^{34,35} this may arise due to the infiltration of hydrogen atoms into the niobium metal before the formation of the surface oxide or during nanofabrication processes such as wet etching. Interestingly, the H⁻ counts are found to be an order of magnitude higher in the uncapped film compared with that in the Ta-capped film.

On the other hand, the F⁻ counts in the Ta oxide are almost an order of magnitude larger than those in the Nb oxide. The observed disparity in F⁻ counts may explain why, in this case, we observe that Ta oxide is roughly 5 nm, whereas previous reports have indicated values closer to 3 nm.^{23,26,27,31,36–38} While it has been reported that the tunneling of H⁻ ions has been proposed as a loss mechanism in the GHz range,^{39,40} it is worth noting that the F⁻ ions are significantly larger than H⁻ ions, and their impact on loss requires further investigation.

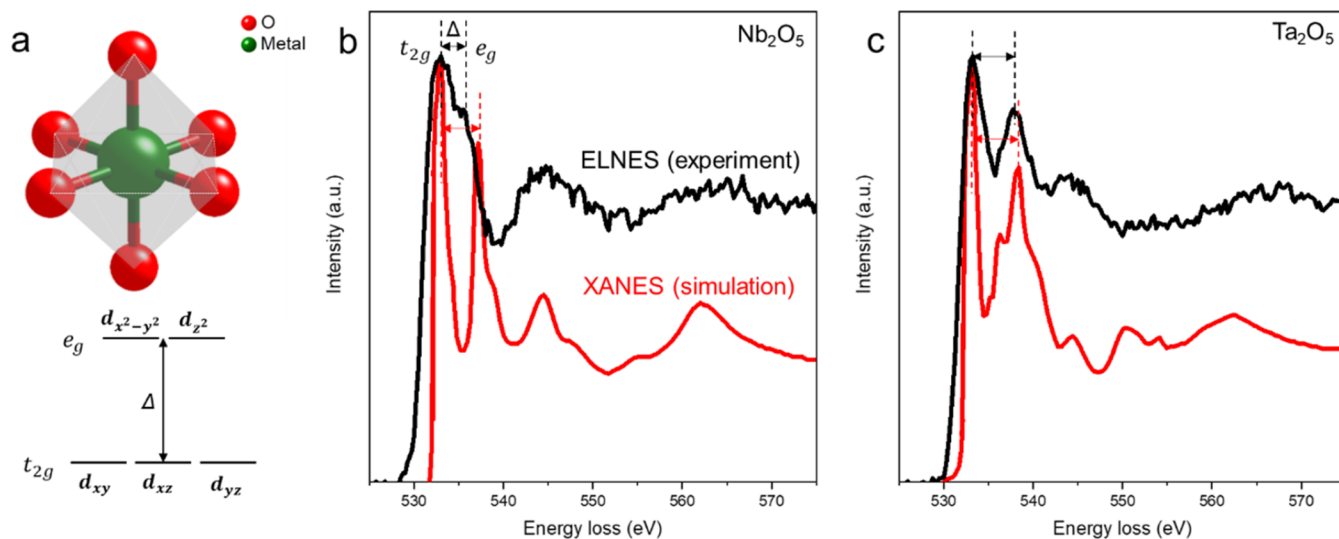


Figure 6. ELNES of O K edge for Nb_2O_5 and Ta_2O_5 . a, Schematic of the octahedrally coordinated transition metal ion, causing a splitting Δ in the energy levels of the d -orbitals. b, O K edges of integrated experimental ELNES of amorphous Nb_2O_5 and simulated XANES of crystalline Nb_2O_5 . c, O K edges of integrated experimental ELNES of amorphous Ta_2O_5 and simulated XANES of crystalline Ta_2O_5 .

The experimentally observed abrupt transition in oxidation states from Ta_2O_5 to Ta in contrast to the gradual shift from Nb_2O_5 to Nb is consistent with the known thermodynamics. Thermodynamic descriptions of the Ta–O and Nb–O systems were obtained using the CALculation of PHase Diagrams (CALPHAD) method based on existing databases, and the resulting phase diagrams are shown in Figure 5.^{41–44} The former has a single stable stoichiometric oxide (Ta_2O_5), while the latter includes three oxides (Nb_2O_5 , NbO_2 , and NbO). The existence of a single valence state in the Ta–O system and three valence states in the Nb–O system is explained by the oxygen chemical potentials (μ_{O}) in both systems, which are plotted as functions of the oxygen mole fraction in Figure 5. There is a single discontinuity in μ_{O} in the Ta–O system, corresponding to an abrupt change in the oxidation state. However, in the Nb–O system, three discontinuities exist in μ_{O} , resulting in a gradual change in the oxidation state. Although the thermodynamic analysis here is based on equilibrium crystalline phases from the phase diagrams, the same trend still holds for the amorphous native oxides.

Since suboxide species may act as magnetic impurities and lead to quasiparticle loss,^{27,38,45,46} understanding the underlying origin of enhanced performance in Ta-based qubits is often solely attributed to suboxide species. However, given that the predominant oxide species for both Nb and Ta exist in the form of their pentoxides, and extensive research indicates that Nb_2O_5 is primarily accountable for two-level system (TLS) loss,^{17,28,29} it suggests there might be another mechanism rooted in the intrinsic material properties of amorphous Nb_2O_5 and Ta_2O_5 . This mechanism could also play a crucial role in the performance improvement of Ta-based or Ta-capped qubits.

The unoccupied p-type states of the surrounding oxygen atoms are hybridized with the neighboring transition metal; the O K edge could provide information that reflects the bonding states in transition metal and O bonding and their oxidation states.^{47,48} Figure 6 directly compares the ELNES of the O K edges for Nb_2O_5 and Ta_2O_5 . Figure 6a illustrates an octahedrally coordinated transition metal atom to oxygen

atoms, offering a qualitative explanation for the differences in the double-peak structure of the O K ELNES based on ligand-field theory. This structure results in the splitting of transition metal d orbitals into the upper, e_g orbital, and the slightly lower in energy t_{2g} , with a crystal-field splitting energy Δ .^{49,50} Figure 6b,c presents the O K edges of measured EELS and calculated X-ray absorption spectroscopy (XAS) for Nb_2O_5 and Ta_2O_5 . Calculated XAS for the crystalline references is employed as EELS, essentially equivalent to XAS under high primary energy and low scattering angle approximations.⁴⁷ Figure 6b depicts the amorphous character of Nb_2O_5 . We observe a decrease in crystal-field splitting energy Δ compared with the crystalline counterpart and broader individual peaks. The simulated X-ray absorption near-edge structure (XANES) exhibits an Δ of 4.15 eV, whereas experimental ELNES exhibits an Δ of 2.5 eV, which is about a 40% reduction. This aligns with measured and calculated EELS and XAS in the literature for amorphous Nb_2O_5 .^{48,50}

In comparison, Figure 6c reveals that the crystal-field splitting energy Δ of the measured ELNES and the calculated XAS of Ta_2O_5 are almost identical (XANES with Δ of 5.0 eV and ELNES with Δ of 4.75 eV, which is about a 5% reduction), consistent with experimental and simulated EELS data reported in ref 33. Moreover, the measured double peaks of Ta_2O_5 are sharper than those of Nb_2O_5 , indicating that amorphous Ta_2O_5 retains a more octahedrally symmetric crystalline character.⁴⁸ This implies a reduced likelihood of tunneling between the two lowest-energy states as predicted in disordered materials, resulting in fewer TLS defects being coupled with the qubit's electric field and reduced microwave loss.^{9,51} On the other hand, amorphous Nb_2O_5 , based on the measured ELNES, may have more distorted octahedrons. This distortion could lead to a higher possibility of easier tunneling between two minimum energy states, akin to the unique two-state nature of the transformation known as the umbrella flip.⁵² As deviations from crystalline order can lead to quantum decoherence, such a phenomenon may further limit qubits' coherence time. More generally, TLS-type loss typically arises from local disorder; hence, an amorphous region is more likely to host such loss mechanisms than its crystalline counterparts.

Finally, the less distorted Ta₂O₅ structure may be more effective in blocking the diffusion of H atoms toward the oxide/metal interface (Figure 4c,d), potentially further reducing TLS sources.

CONCLUSIONS

Structural and chemical investigations were conducted on the M-A interface to elucidate the reasons behind coherence time improvement when native TaO_x was utilized instead of native NbO_x. Our observation revealed an abrupt transition from Ta₂O₅ to Ta metal, marked by a significant absence of suboxide species, contrasting with the gradual valence state transition from Nb₂O₅ to Nb metal in native NbO_x. The calculated phase diagram and oxygen chemical potential between Nb–O and Ta–O systems explain this disparity. In addition, we discovered a closer crystalline bonding nature of amorphous Ta₂O₅ than that of Nb₂O₅. We attribute this distinction to another potential origin for the enhanced T₁ of Ta or Ta-capped Nb superconducting qubits. Our comprehensive approach, combining STEM-EELS and thermodynamic calculations, elucidates the formation mechanism and low-loss nature of TaO_x compared to those of NbO_x. This insight addresses current gaps in understanding low-loss TaO_x properties, creating a foundation for subsequent in-depth theoretical investigations to validate these findings further.

METHODS

Qubit Fabrication. The qubit devices were fabricated on high-resistivity Si substrates ($\rho \geq 10$ k Ω -cm) with sputter-deposited Nb (~200 nm thick) or Ta-capped (~10 nm) Nb layers and Al/AlO_x/Al Josephson junctions. Details on the thin film and device fabrication have been reported elsewhere.²³

AFM Analysis. The surface morphology of the Nb- and Ta-capped Nb films was analyzed by AFM using an Asylum Cypher AFM with a Si cantilever in tapping mode at a scanning rate of 1 Hz. The image resolution was 512 × 512 pixels.

(S)TEM-EELS Analysis. Cross-sectional TEM samples were prepared by using a focused ion beam instrument with a gas injection system (Helios, Thermo Fisher Scientific Ltd.). The TEM samples were thinned to electron beam transparency by a Ga⁺ ion beam from 30 to 2 kV. The TEM samples were investigated using an aberration-corrected TEM (Titan Cube, Thermo Fisher Scientific Ltd.) at 200 kV. A high-angle annular dark-field (HAADF) detector was used for dark-field imaging in STEM mode with a convergent semiangle and a collection semiangle of 18 and 74–200 mrad, respectively. Energy-dispersive X-ray spectroscopy (EDS) and electron energy-loss spectroscopy (EELS) studies were carried out with probe currents of 200 and 50 pA, respectively. Dual EELS was performed to acquire all-electron energy-loss (ELL) spectra. The plural scattering effect of all of the raw ELL spectra was removed via Fourier-ratio deconvolution.

XPS Analysis. XPS analysis was conducted with a Kratos Amicus/ESCA 3400 device, utilizing 240 W Mg K α X-rays from a twin-anode laboratory source. The emitted photoelectrons were collected at a perpendicular angle to the surface and analyzed using a DuPont-type analyzer, with the pass energy adjusted to 150 eV. To refine the spectra, a Shirley baseline correction was applied using the CasaXPS software, and charging effects were compensated by calibrating the C 1s peak binding energy to 285 eV.

ToF-SIMS Analysis. ToF-SIMS measurements were performed using a dual beam IONTOF 5 system to analyze impurities' concentration and depth distribution in the Nb and Ta/Nb films. Secondary ion measurements were performed by using a liquid bismuth ion beam (Bi⁺). A cesium ion gun with an energy of 500 eV sputtered the surface for depth profile measurements. All counts were

normalized to the total counts in the Nb metal layer, enabling qualitative comparison of impurities between samples.

XAS Simulation. Simulation of X-ray absorption spectra was created by Materials Project,⁵³ for orthorhombic Nb₂O₅⁵⁴ and monoclinic Ta₂O₅.⁵⁵

Thermodynamic Calculations. Thermodynamic models of the Ta–O and Nb–O systems using the CALPHAD method were adopted from a prior study⁴¹ and from the TCOX11 database⁴⁴ in the Thermo-Calc software.⁴³ The Ta–O models include low-temperature orthorhombic (β) and high-temperature tetragonal (α) Ta₂O₅ polymorphs, body-centered cubic (BCC) α -Ta, liquid, and gas phases, which were respectively described as stoichiometric compounds, a solid solution, ionic liquid,⁵⁶ and ideal gas. The Nb–O models, based on unpublished work by Dupin and Ansara⁵⁷ and published by Massih and Pérez,⁵⁸ include monoclinic Nb₂O₅, tetragonal NbO₂, cubic NbO, BCC Nb, liquid, and gas phases. The compounds were modeled as stoichiometric, BCC Nb as a solid solution, the liquid as an ionic liquid, and the gas as ideal.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsnano.4c05251>.

Principal component analysis (PCA) of O K edge EELS spectra across the TaO_x-Ta interface (PDF)

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Notes

The authors declare no competing financial interest.

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After this paper was published ASAP July 22, 2024, we corrected a spelling error in the name of author Carlos G. Torres Castanedo. The corrected version was reposted July 30, 2024.