Supplementary Material for "Characterization and reduction of microfabrication-induced decoherence in superconducting quantum circuits"

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I. FABRICATION OF $\lambda/2$ LIFT-OFF RESONATORS

We describe in greater detail the fabrication process for the resonators of Fig. 1 of the main text. First, a polished *c*-plane sapphire wafer is solvent-cleaned (5 min. sonication in acetone and then isopropanol [IPA], spun dry) and a 100 nm base layer of aluminum is deposited in a high vacuum *Plassys* electron-beam evaporator (base pressure $\sim 5 \times 10^{-8}$ Torr) after a short and gentle *in situ* argon ion beam clean (beam energy $200\,\mathrm{eV}$, dose $\sim 5 \times 10^{15}\,\mathrm{cm}^{-2}$). The photolithographic etch used to form the control resonators and to etch the entire ground plane slot of the lift-off resonators was a BCl_3/Cl_2 inductively coupled plasma (ICP) etch with a subsequent quench with CF_4 and then DI water.

The center traces for the lift-off resonators are fabricated as follows: the wafer is solvent-cleaned and then dehydration-baked (3 to 4 min. on a hotplate at $115\,^{\circ}\mathrm{C}$) and allowed to cool, after which a bilayer of 950K PMMA (300 nm) atop copolymer MMA(8.5)MAA (500 nm) e-beam resists are spun, each baked at $160\,^{\circ}\mathrm{C}$ on a hotplate for 10 minutes. The center traces are then defined with electron-beam lithography ($100\,\mathrm{kV}$, $2\,\mathrm{nA}$ beam in a JEOL JBX-6300FS electron beam writer, with a dose of $2000\,\mu\mathrm{C/cm}^2$), after which the bilayer is developed with various development times in a 1:3 mixture of methyl isobutyl ketone (MIBK) to isopropanol (IPA), followed by a 10 second IPA dip and thorough nitrogen blow-dry. It is possible that the IPA dip encouraged re-deposition of resist onto the substrate, but we did not test this.

After development, the surface is optionally treated with a downstream oxygen ash descum before center trace deposition. During this descum, the substrate is heated to $150\,^{\circ}\mathrm{C}$ and sees purely chemical cleaning with reactive oxygen, but not ions or plasma (3 min. in a Gasonics Aura 2000-LL downstream asher. We note that we found it necessary to pre-clean the tool's chamber with a stronger recipe to obtain consistent descum results at the relatively low $150\,^{\circ}\mathrm{C}$). The wafer is then transferred to and pumped overnight in the same *Plassys* high-vacuum e-beam evaporator used for the initial ground plane deposition, and the 100 nm Al center traces are then deposited for the lift-off resonators without any *in situ* clean. The excess metal is then lifted off in N-Methyl-2-pyrrolidone (NMP) at $80\,^{\circ}\mathrm{C}$ (3 hrs.), sonicated in NMP then IPA, and then spun dry.

II. OBSERVED FEATURES WITH NEGLIGIBLE LOSS CONTRIBUTIONS

Resonator edge profiles [Fig 1(c/d) of main text]: the sidewall of the etched metal is nearly vertical, with slight etching on the metal due to resist delamination, whereas the lift-off metal sidewall has an angle of $\sim 25^{\circ}$ from the vertical.

Substrate roughness: the substrate under the center trace of the lift-off resonators previously saw an etch that increased the substrate roughness to 0.3 from 0.1 nm RMS roughness (the substrate roughness under the center trace of the control resonators) as measured with atomic force microscopy.

Residual granules: For the lift-off resonators without descum, atomic force microscope (AFM) scans of the substrate after e-beam resist exposure and development also reveal residual resist granules with widths of $\sim 10-100\,\mathrm{nm}$ and heights $\sim 2-20\,\mathrm{nm}$, even significantly above the e-beam clearing dose exposure, consistent with literature on PMMA.[1] However, we expect these granules to be negligible sources of dielectric loss compared with residual films, as the space they fill is negligible (areal fraction $\sim 0.5\,\%$ and equivalent uniform thickness $\lesssim 0.1\,\mathrm{nm}$). Downstream ashing or UV-Ozone cleaning mostly removes these granules. We note that unlike some direct oxygen plasma treatments, these treatments could also be used on a silicon substrate without oxidizing it beyond native oxide levels.

III. TEM SAMPLE PREPARATION AND IMAGING

The TEM sample of Fig. 1 contains S-M interfaces that are similar, but not identical, to those of the center traces of the lift-off resonators without/with the descum. Differences arise due to fabricating multiple interfaces on a single TEM sample: The non-ashed S-M interface in the TEM sample saw processing temperatures up to $160\,^{\circ}\mathrm{C}$, whereas the ashed S-M interface in the TEM sample and in the resonators only saw processing temperatures up to $115\,^{\circ}\mathrm{C}$. As such, it is not certain if the upper AlO_x sublayer was present in the non-ashed lift-off resonators. In addition, the ashed S-M interface in the TEM sample saw an initial coating of e-beam resist and subsequent strip before a second coating for e-beam lithography.

The electron-transparent TEM samples for Fig. 1 and

Fig. 2 were $50-100$ nm thick, prepared via a gallium focused ion beam lift-out technique using protective coatings of Ir and Pt. For HRTEM images, the electron beam (200 kV) was aligned with sapphire substrate low-index zone axis to ensure the that interfacial layers were viewed edge-on. Images shown in the main text are phase contrast images. The difference between the shades of the substrate between Fig. 2(b) and 2(c) is mainly differing diffraction contrast between two different samples.

The samples were plasma cleaned prior to EELS analysis to reduce any background surface contamination. EELS elemental analysis has a spatial resolution of $1-2$ nm, but was taken at an angle across the interfaces to increase sampling and effective resolution within the interface. We do not extract any quantitative data from the EELS scans, but they allow us to see the clear presence or absence of local peaks in the core-loss signal corresponding to different elements of moderately low atomic number. In particular, no signal for B, Cl, or F (elements associated with the ICP etch) was detected for the samples in Fig. 1 of the main text.

IV. ION MILL PARAMETERS AND INDUCED LOSS

The *in situ* pre-deposition argon ion milling steps were performed in the *Plassys* high-vacuum e-beam evaporator using a Kaufman source with a ~ 3 inch beam diameter. The parameters for the weak ion beam treatment are beam energy 200 eV and current ~ 4 mA for 10 seconds, for a dose of $\sim 5\times10^{15}\,\mathrm{cm}^{-2}$. The parameters of the stronger ion beam treatment are beam energy 400 eV and current ~ 20 mA for 3.5 minutes, for a dose of $\sim 5\times10^{17}\,\mathrm{cm}^{-2}$. We did not measure resonators whose substrates saw no ion beam treatment whatsoever.

The aluminum ICP dry etch used to define the resonators etches ~ 5 nm into the sapphire substrate. Accordingly, when we extract a loss tangent for the interfacial layer created by the strong ion mill [Fig. 2(c) of the main text], we only attribute participation to this layer at the S-M interface and not the partially etched S-V interface.

V. NATIVE OXIDE IN THE TRAP CAPACITOR EXPERIMENT

The passivating native oxide in the trap capacitor [left panel of Fig. 3 of the main text] was originally grown by oxidizing the ground plane at 5 mbar in the *Plassys* high-vacuum evaporator immediately after deposition. A separate experiment in which the residue in the trap capacitor was partially removed from the native oxide via a short descum (only a 1 min. downstream ash was used) lets us estimate an upper bound of $\sim 2\times10^{-3}$ for δ^0_{TLS} of the native oxide at ~ 1 V/m, though it is possible[2] that going to even lower powers would reveal a second, higher δ^0_{TLS}. The thickness range of this oxide quoted in the main text was extracted by resistance measurements, TEM, and ellipsometry. It would be interesting to repeat this experiment to test other kinds of contamination.

VI. BULK RESIST LOSS TANGENT EXPERIMENTS

For the experiment in which we coated resonators with 500 nm of PMMA copolymer e-beam resist, when we repeated the measurements after exposing the resist, we exposed for 50 minutes in a DUV flood exposer, for a dose of $\sim 60\,\mathrm{J/cm^2}$ with wavelengths between 200 and 260 nm. These resonators were on a high-resistivity silicon substrate rather than sapphire, for ease of exposing the edge of the die with a scanning electron microscope for the sake of clearing the edges for subsequent wirebond connections to the resist-coated chip. These resonators were measured near 4, 5, and 6 GHz, with geometries of $W/G = 3/2$, $6/4$ and $15/10$ μm. The extracted bulk resist loss tangent and dielectric constant was the same for all three frequencies to within experimental uncertainty (as quoted in the main text), and the loss scaled very closely with the electric energy filling factor of the resist according to COMSOL simulations. As a general note used for the calculations in this work, for computing resonator quality factors from the participation of lossy dielectrics, one must remember to account for the stored inductive energy in the resonator as well.

VII. PLASMA ETCH POST-TREATMENT

We post-treated resonators in a Technics PE-IIA oxygen plasma etch system (300 mT O_2, 100 W power for 30 sec., a common cleanroom "descum" recipe). As mentioned in the main text, this decreased low-power Q_i down to $\sim 200{,}000$. It also pushed the power at which high-power Q_i saturates to above the power at which the resonators show a nonlinear response. This is consistent with the addition of TLS with a relatively high saturation field, or very numerous or lossy TLS in a region that does not participate strongly (i.e., the M-V interface); however, we have not yet characterized the origin of loss induced by exposure to this aggressive oxygen plasma. There was no observed frequency shift in the resonators due to this treatment.

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