Engineering Nonlinear Response of Superconducting Niobium Microstrip Resonators via Aluminum Cladding

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In this work, we find that Al cladding on Nb microstrip resonators is an efficient way to suppress nonlinear responses induced by local Joule heating, resulting in improved microwave power handling capability. This improvement is likely due to the proximity effect between the Al and the Nb layers. The proximity effect is found to be controllable by tuning the thickness of the Al layer. We show that improving the film quality is also helpful as it enhances the microwave critical current density, but it cannot eliminate the local heating.

I. INTRODUCTION

Recently, superconducting planar resonators1 have found applications in magnetic resonance because their low dissipation and small mode volume have greatly improved the sensitivity of spin detection.2–11 In addition to the detection sensitivity, superconducting resonators are required to handle strong microwave pulses for efficient spin manipulation. However, microwave power handling capability of superconducting planar resonators has been an issue—if we apply a strong microwave pulse, the nonlinearity of the resonator participates such that the actual microwave magnetic field that a spin sees becomes significantly different from what we intended.

Regarding the Duffing-type nonlinearity12, it is known that we can design a pulse which can compensate the nonlinear response as the Duffing-type nonlinearity can be easily modeled and controlled using nonlinear circuit models.13–18 However, many reported nonlinear responses of type-II superconducting resonators are very difficult to model, thus not controllable: the shape of S-parameter curves becomes irregular and greatly suppressed even at modest microwave power. This type of nonlinearity has been attributed to local Joule heating, often called a hot spot19,20–29 followed by switching of weak links to the normal state.15,20–29 Thus, it is crucial to minimize this undesired nonlinear response for magnetic resonance applications.

In this work, we investigated ways to suppress the nonlinearity due to local Joule heating by improving the film quality (Sec. III) and Al cladding (Sec. IV). We first showed that local Joule heating is the dominant source of the nonlinearity of pure Nb microstrip resonators. Then, we found that a resonator made of better quality film showed a significantly higher microwave critical current, but the major mechanism of the nonlinearity remains the same. Meanwhile, Al cladding effectively eliminated the nonlinear responses induced by local Joule heating. This improved microwave power handling capability is likely due to the proximity effect between the Al and the Nb layers.31–34 The existence of the proximity effect was confirmed experimentally by studying how magnetic field dependence of the resonance frequency f and the quality factor Q change as we tune the thickness of the Al layer (Sec. V). This study also showed that the proximity effect is controllable by tuning the thickness of the Al layer.

II. METHODS

Four microstrip resonators with different film quality and Al cladding thickness were used. Two of them were pure Nb resonators with different film quality. The film quality was controlled mainly by the temperature of the substrate at the time of the film growth; the higher substrate temperature resulted in higher critical temperature and lower residual resistivity, i.e., a better quality film. The other two resonators were trilayer resonators—Nb resonators with Al cladding. For all resonators, the ground plane was made of a pure Nb layer, and the thickness of Nb layers, including the ground plane, was 50 nm. Since our resonators are all microstrip resonators, i.e., double-sided film, there is no complication for making the ground plane and the microstrip with different material compositions.

The resonators are labeled with the thickness of the

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Therefore, we believe a microstrip geometry is more suitable for electron spin resonance (ESR) of thin films, which is our research interest\textsuperscript{[21,39]} As a trade-off, the less confined field profile inherently leads to more radiation loss and dielectric loss; thus, our resonators show lower internal quality factor (see Table \textbf{IV}) values than corresponding coplanar resonators.

The measurements were made in a cryogen-free di-lution refrigerator (Leiden CF250). The resonator is aligned with a magnetic field using a goniometer (At-tocube ANGt101) with the precision \(\pm 5\) mdeg at 0.1 K. For more details, see Sec. III of Ref. \textsuperscript{35}

Full S-parameters were collected using a vector network analyzer (VNA, Agilent N5230A). The resonance frequency and the loaded quality factor \(Q_{\text{load}}\) were obtained by fitting the magnitude of the measured \(S_{21}\) to a complex Lorentzian function\textsuperscript{[35]}. The external quality factor \(Q_{\text{ex}}\) was obtained using the formula \(Q_{\text{load}} = Q_{\text{ex}}e^{-10IL/20}\), where IL is the insertion loss in dB (Ref. \textsuperscript{36}). The internal quality factor \(Q_{\text{in}}\) was obtained from the relation \(Q_{\text{in}}^{-1} = Q_{\text{ex}}^{-1} + Q_{\text{circ}}^{-1}\). The input microwave power \(P_{\text{in}}\) was estimated by \(P_{\text{in}} = P_{\text{VNA}} - \text{CL}\) (in dB), where \(P_{\text{VNA}}\) is the setting power of the VNA, and CL is the loss in cables from the VNA to the input capacitor of the resonator\textsuperscript{1}. The circulating power \(P_{\text{circ}}\) was estimated using \(P_{\text{circ}} = \pi^{-1}P_{\text{in}}Q_{\text{load}}^{-1}e^{-10IL/20}\). The maximum value of the microwave current \(I_{\text{circ}}\) circulating in the resonator was estimated using \(I_{\text{circ}} = \sqrt{8P_{\text{circ}}/Z_0}\) (Ref. \textsuperscript{37}), where \(Z_0\) is the characteristic impedance of the resonator, which is designed to be about 50 $\Omega$.

### III. IMPROVING THE FILM QUALITY

Figure \textsuperscript{2}(a) shows \(S_{21}\) curves of Al-0L at various \(P_{\text{in}}\). As \(P_{\text{in}}\) increases, the resonance peak becomes rapidly suppressed and irregular. To know the source of this non-linearity, we plotted the maximum value of \(S_{21}\) (\(S_{21}^{\text{max}}\)) as a function of \(P_{\text{in}}\) [Fig. \textsuperscript{2}(c)]. Note that the plot can be divided into three regions based on the shape of the \(S_{21}\) curve: In the low-power region (region I), the \(S_{21}\) curve is Lorentzian. As \(P_{\text{in}}\) increases (region II), the curve becomes asymmetric and distorted. Finally, the resonator enters region III with the onset of jump in \(S_{21}^{\text{max}}\). In this region, the shape of the \(S_{21}\) curve is very irregular; when the curves were swept in the opposite direction, the \(S_{21}\) curve becomes reflected shape [dashed lines in Fig. \textsuperscript{2}(a)].

The same measurements were done with a resonator made of a better quality Nb film, Al-0H [Fig. \textsuperscript{2}(b)]. There are several notable differences between Al-0L and Al-0H: First, for Al-0H, the shape of the \(S_{21}\) curve is Lorentzian and the resonance frequency remains the same until \(P_{\text{in}}\) reaches \(-21\) dBm. However, there is an abrupt drop in

- The estimated insertion loss can vary 1–2 dB from one package to the next mainly due to imperfect package assembling. Such a deviation is not crucial for the power dependence, but it may change the values of the internal quality factor in Table \textbf{IV} about 20–30%.

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|}
\hline
Res. & Composition & \(T_{\text{growth}}\) & \(f_0\) (GHz) & \(Q_0\) \\
\hline
Al-0L & Nb 50 & low & 10.0728 & 14300 \\
Al-5L & Al5/Nb 50/Al5 & low & 9.9764 & 21000 \\
Al-10L & Al10/Nb 50/Al10 & low & 10.0672 & 23600 \\
Al-0H & Nb 50 & high & 10.0792 & 27500 \\
\hline
\end{tabular}
\caption{The name convention of resonators and their resonance frequency \(f_0\) and loaded quality factor \(Q_0\).}
\end{table}

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{figure1}
\caption{Geometry of the resonators. \(G\) is the gap between the feedline and the resonator. \(W\) is the width of a strip. \(S\) is the spacing between center of strips. The value of \(G\) is 400 for Al-0H and 350 $\mu$m for other resonators; \(W\), 15 $\mu$m; and \(S\), 75 $\mu$m. The distance between the strips and the ground plane is 430 $\mu$m. For clarity, the ground plane is not shown.}
\end{figure}
TABLE II. Summary of the Nb film growth conditions. The two rightmost columns are resulting critical temperature $T_c$ and residual resistivity $\rho_n$ from transport measurements. RT stands for Room Temperature. “Ar sputter cleaning” means Ar sputter cleaning of the substrate, i.e., back sputtering, before the film growth. For further details about the growth conditions, see Ref. 30 and the supplementary material of Ref. 35, in which the films for Al-0H and Al-0L are appeared as wafers A and C, respectively. For more details about the resulting film properties, see Table I of Ref. 35.

| Res. | Base pressure (mbar) | Growth rate (Å/s) | Ar pressure (mbar) | Temperature: strip (°C) | Temperature: ground plane (°C) | Ar-sputter cleaning | $T_c$ (K) | $\rho_n$ (µΩ cm) |
|------|---------------------|-------------------|-------------------|-------------------------|-------------------------------|---------------------|-----------|-----------------|
| Al-0L | $\sim 10^{-8}$ | 0.6 | $4 \times 10^{-3}$ | RT | RT | Yes | 7.2 | 17 |
| Al-0H | $\sim 10^{-10}$ | 1.7 | $2 \times 10^{-3}$ | 550 | 770 | No | 9.3 | 2.9 |

$S_{21}^{max}$ and the quality factor near $P_{in} = -29$ dBm; hence, we divide region I into regions Ia and Ib. The reason for this sudden drop is unclear at this stage.

Secondly, in region III, Al-0H shows cratered Lorentzian shapes and bistability, which have been accounted for switching of weak links, such as grain boundaries, to the normal state. For Al-0L, the irregular shape is reproducible from sweep to sweep—there is no notable bistable behavior. This suggests that the spread in the microwave critical currents of Al-0L more significant than that of Al-0H such that bistable behaviors are averaged out, resulting in spike-like features (see Fig. 9 in Ref. 27).

For quantitative understanding, we estimated microwave current densities at the boundaries between regions; the values are summarized in Table III. Here, note that, $j_{\text{microwave}}^{II,III}$, which can be considered as the microwave current density, is defined by the maximum $S_{21}$ in the crater, as denoted by arrows in (a,b). The data were taken at zero field and about 0.2 K.
stronger than that between Al and Al. The reason for this is that the bonding between Al and Nb is temperature wet on the surface of Nb ideally. The Al-Nb combination is special because Al grown at room temperature wets on the surface of Nb perfectly. In this regard, Al layers need to cover the field of the Al layers are substantially enhanced. Here, to provide reliable bypasses, the Al layers need to cover the surface of the Nb layers perfectly. In this regard, Al and Nb combination is special because Al grown at room temperature wets on the surface of Nb ideally. (see Sec. I) and simulations for the current density distribution. $j^{GL}_{i,j}$ was calculated using $j^{GL}_{i,j} = (2/3)^{1.5} H_i / \lambda$ (Ref. 55). For Al-0L, $\mu_0 H_i$ and $\lambda$ were taken from Table IV for Al-0H, 0.27 T and 52 nm, respectively. The units are dBm for circulating powers and $A/m^2$ for current densities.

| Res.  | $P_{c,circ}^{I,II}$ | $P_{circ}^{II,III}$ | $j_{mw}^{I,II,III}$ | $j_{mw}^{II,III}$ | $j_{d,GL}^{I,II,III}$ |
|-------|-------------------|--------------------|-------------------|-----------------|---------------------|
| Al-0L | 1.2               | 4                  | $1.5 \times 10^{10}$ | $2 \times 10^{10}$ | $5 \times 10^{11}$  |
| Al-0H | 11                | 18                 | $1.3 \times 10^{11}$ | $3 \times 10^{11}$ | $2 \times 10^{12}$  |

critical current density, is about one order of magnitude less than $j_{d,GL}$ for both resonators, suggesting that the microwave power handling capability is not limited by intrinsic and global properties of superconductivity. This supports that the dominant mechanism for the nonlinearity of pure Nb films is local Joule heating. Also note that $j_{mw}^{II,III}$ of the resonator made of a better quality film (Al-0H) are about one order of magnitude higher than that of the resonator with low film quality (Al-0L). This shows that improving the film quality enhances the microwave critical current density, but it does not change the dominant mechanism for the nonlinearity.

IV. ALUMINUM CLADDING

Figure 3 shows $S_{21}$ curves at various $P_n$ after Al cladding. Note that Al cladding changes the nonlinear response dramatically: Al-5L and Al-10L show the Duffing-type nonlinearity instead of irregular shapes. Since this type of nonlinearity is controllable using nonlinear circuit model, we can say that Al cladding improves the high-power handling capability.

The qualitative change in the nonlinearity after Al cladding is likely due to current bypasses provided by Al near weak links in the Nb layer. The superconductivity of these Al bypasses is strengthened by the proximity effect such that the critical current density and the critical field of the Al layers are substantially enhanced. Here, to provide reliable bypasses, the Al layers need to cover the surface of the Nb layers perfectly. In this regard, Al and Nb combination is special because Al grown at room temperature wets on the surface of Nb ideally. (see Sec. I) and simulations for the current density distribution. $j^{GL}_{i,j}$ was calculated using $j^{GL}_{i,j} = (2/3)^{1.5} H_i / \lambda$ (Ref. 55). For Al-0L, $\mu_0 H_i$ and $\lambda$ were taken from Table IV for Al-0H, 0.27 T and 52 nm, respectively. The units are dBm for circulating powers and $A/m^2$ for current densities.

Other possible roles of Al cladding, such as protection against oxidation of the Nb layer and enhancing the thermal conductivity can reduce the number of weak links and local Joule heating; however, they cannot fully account for such a qualitative change. Here, note that a normal metal layer can do the same things, except strengthening the superconductivity. In Refs. 26 and 27, a 35 nm thick layer of Au was deposited on an MgB$_2$ thin film, but the nonlinear behavior due to switching of weak links remained largely unchanged. In addition, another way to reducing the number of weak links, improving the film quality, does not change the dominant mechanism for the nonlinearity as shown in Sec. III. From these results, we believe that the qualitative change in nonlinear response after Al cladding is mainly due to the proximity effect.

Even after the switching of weak links are eliminated, further Al cladding can still assist, as shown in Fig. 3(b). Note that, in Table IV, Al-10L shows higher internal quality factor ($Q_{0,ini}$) which implies that Al cladding reduces the surface resistance, and lower effective residual resistivity ($\rho_{ini}$) compared to Al-5L. (See Sec. V for further explanation regarding Table IV). These results suggest that the observed nonlinearity of the trilayer resonators is driven by global heating which is generated through the following process: When the circulating power is low enough such that there is no notable
nonlinear response, the heat balance between the cooling power and the dissipated power due to the finite surface resistance is fulfilled. At this stage, there are not many thermally-excited quasiparticles because of the low temperature (0.2 K). When the circulating power passes a certain level, at which the dissipated power is greater than the cooling power, quasiparticles are excited and participate in the power dissipation, which is proportional to $\rho_n j_{\text{diss}}^2$, where $\rho_n$ is the residual resistivity.

The intrinsic GL nonlinearity does not account for the nonlinearity of the trilayer resonators because the GL nonlinearity is known to be much more reactive than shown by the data in Fig. 4 (Ref. 50). Indeed the data in Fig. 4 of Sec. V, which follow the GL equations closely, the shift of $f$ about 1 MHz does not result in notable change in $Q$ and $S_{21}^{\text{max}}$. In addition, vortex penetration into grains is also unlikely because, if vortices were created by a microwave current and penetrated into the grains, a hysteretic behavior would be observed due to vortex pinning; in other words, the $S_{21}$ curve would not go back to its original position and shape once high $P_{\text{in}}$ was applied. Such behavior was not observed at zero field for all resonators.

In a modest field parallel to the microwave current $H_{||}$, we find that the results in Figs. 2 and 3 are largely unchanged. Some of the representative data are shown in Fig. S3. In a field perpendicular to the film $H_{\perp}$, we found that applying a high microwave current results in magnetic hysteresis caused by suppression of the edge barrier and consequent injection of vortices. Here, these vortices are created by $H_{\perp}$, not by the microwave current. The supporting data and analysis are in Sec. S3.

V. THE PROXIMITY EFFECT

In this section, we experimentally confirm the existence of the proximity effect between the Al and the Nb layers and show this effect is controllable by varying the thickness of the Al layer. For this, we investigate how a magnetic field parallel to the microwave current $H_{||}$ dependence of $f$ and $Q$ change as we tune the thickness of the Al layer. Figure 4 shows how the thickness of the Al layers affects the $H_{||}$ dependence of $Q$. Note that, as the thickness of the Al layers increases, $Q$ starts to drop at a lower field, which already indicates the existence of the proximity effect.

In addition, as already mentioned in Sec. IV, $Q_{0,\text{in}}$ becomes higher as the thickness of the Al layers increases. The origin of higher $Q_{0,\text{in}}$ after Al cladding is probably less-lossy surface oxide or an improved interface between the substrate and the film. Thermal quasiparticles are not relevant because, at the measurement temperature ($\leq 20$ mK), thermal quasiparticles are expected to be frozen out.

For quantitative analysis of the proximity effect, we characterize the $H_{||}$ dependence of $f^{-2}$ and $Q$ in Fig. 4 using a set of parameters, which we call loss parameters, associated with magnetic field induced quasiparticle generation. The basis of this approach is that the magnetic field dependence of the real and imaginary parts of the complex resistivity $\rho_1 + i\rho_2$ can be studied via $Q$ and $f$ as a function of field, respectively. We emphasize that these loss parameters, shown in Table IV, were obtained purely by comparing the measured and expected $f$ and $Q$ without incorporating any other types of measurements. To calculate the expected $f$ and $Q$ as a function of $H_{||}$, we need to model the complex resistivity.

In order to model the complex resistivity as-
TABLE IV. Loss parameters extracted from Fig. 4 by following the procedures described in Sec. S2. The internal quality factor below 20 mK without a magnetic field $Q_{0,\text{in}}$ and the external quality factor $Q_{\text{ex}}$ are also shown. Here, $Q_{\text{ex}}$ of the resonators in this table are almost the same because the gap between the feedline and the resonator ($G$ in Fig. 4) are designed to be identical for straightforward comparison of the quality factors. $\lambda_0^\parallel$ is the zero-field in-plane penetration depth; $\gamma$ is the anisotropy parameter; $\kappa_\parallel$ is the in-plane GL parameter; $H_c$ is the thermodynamic critical field; $Q_{0,\text{fit}}$ is the zero-field loaded quality factor determined by fitting; $\beta$ is the exponent for the fraction of normal electrons in the context of the two-fluid picture (see Sec. S2 for the formal definition); and $\rho_{n,\text{fit}}$ is the residual resistivity obtained from fitting. The loss parameters of Al-0L are from Ref. 35.

Note that, given the data in Fig. 4, $\rho_{n,\text{fit}}$ cannot be determined independently; any combination of $\kappa_\parallel$ and $\gamma$ gives similar results if $\gamma\kappa_\parallel$ is the same. The reason is that $H_{0,p}$ is roughly proportional to $\xi_\parallel\xi_\perp$ (Ref. 53), and $\xi_\parallel\xi_\perp$ is proportional to $\gamma\kappa_\parallel$.

| Res. | $Q_{\text{ex}}$ | $Q_{0,\text{in}}$ | $\lambda_0^\parallel$ (nm) | $\gamma\kappa_\parallel$ | $\mu_0H_c$ (mT) | $Q_{0,\text{fit}}$ | $\beta$ | $\rho_{n,\text{fit}}$ ($\mu\Omega\cdot\text{cm}$) |
|------|----------------|-----------------|-----------------|-----------------|-----------------|----------------|--------|-----------------|
| Al-10L | $4 \times 10^4$ | $6 \times 10^3$ | 85 | 5.3 | 77 | 2.36 $\times 10^4$ | 1.1 | 4.3 |
| Al-5L  | $4 \times 10^4$ | $5 \times 10^3$ | 200 | 12.2 | 102 | 2.10 $\times 10^4$ | 2.2 | 14 |
| Al-0L  | $4 \times 10^4$ | $2 \times 10^3$ | 162 | 6.5 | 190 | 1.46 $\times 10^4$ | 2.2 | 17 |

TABLE IV. Loss parameters extracted from Fig. 4 by following the procedures described in Sec. S2. The internal quality factor below 20 mK without a magnetic field $Q_{0,\text{in}}$ and the external quality factor $Q_{\text{ex}}$ are also shown. Here, $Q_{\text{ex}}$ of the resonators in this table are almost the same because the gap between the feedline and the resonator ($G$ in Fig. 4) are designed to be identical for straightforward comparison of the quality factors. $\lambda_0^\parallel$ is the zero-field in-plane penetration depth; $\gamma$ is the anisotropy parameter; $\kappa_\parallel$ is the in-plane GL parameter; $H_c$ is the thermodynamic critical field; $Q_{0,\text{fit}}$ is the zero-field loaded quality factor determined by fitting; $\beta$ is the exponent for the fraction of normal electrons in the context of the two-fluid picture (see Sec. S2 for the formal definition); and $\rho_{n,\text{fit}}$ is the residual resistivity obtained from fitting. The loss parameters of Al-0L are from Ref. 35. Note that, given the data in Fig. 4, $\kappa_\parallel$ and $\gamma$ cannot be determined independently; any combination of $\kappa_\parallel$ and $\gamma$ gives similar results if $\gamma\kappa_\parallel$ is the same. The reason is that $H_{0,p}$ is roughly proportional to $\xi_\parallel\xi_\perp$ (Ref. 53), and $\xi_\parallel\xi_\perp$ is proportional to $\gamma\kappa_\parallel$.

The measured data and calculated curves agree well (Fig. 3). This suggests that our system can be treated as a single system with effective parameters, and we should not strongly separate Nb and Al layers in our system. The reason for this is that the Al thickness is well below the coherence length of both Nb and Al. If there is a phase transition from superconducting Al/superconducting Nb to normal Al/superconducting Nb, then there must be some abrupt change in the $H_{\parallel}$ dependence other than vortex injection, but no such a signature was observed.

In Table IV, $H_c$ and $\rho_{n,\text{fit}}$ decrease as the Al thickness increases. This result is consistent with previous reports.\textsuperscript{21,23} Note that $\lambda_0^\parallel$ of Al-5L is longer than that of Al-0L, although the proximity effect is expected to reduce $\lambda_0^\parallel$ (Refs. 51 and 52). This elongation of $\lambda_0^\parallel$ is likely due to electron scattering at the interface. As the Al layer becomes thicker, the contribution of the Al layer to $\lambda_0^\parallel$ becomes dominant compared with the interface. As a result, $\lambda_0^\parallel$ of Al-10L is significantly shorter than that of Al-5L.

Note that, in Table IV, $f_0$ of Al-5L is about 0.1 GHz lower than that of Al-0L. In addition, $f_0$ of Al-10L is 0.1 GHz higher than that of Al-5L. Since $f_0$ is proportional to $1/\sqrt{L}$, where $L$ is the effective inductance per unit length, 0.1 GHz change in $f_0$ means 2% change in $L$. In our geometry, the dominant contribution to $L$ is the inductance from the energy stored as an electromagnetic field $L_{\text{field}}$. From the simulation,\textsuperscript{33} we obtain $L_{\text{field}} \approx 510$ nH/m, suggesting that 0.1 GHz change in $f_0$ corresponds to about 10 nH/m change in $L$. The kinetic inductance per unit length $L_{K1}$ can be calculated using the following formula:\textsuperscript{35,25,26}

$$L_{K1} \approx \mu_0\lambda^2 \int \frac{|J_{\text{mwa}}|^2}{|I|^2} dA,$$

(1)

where $A$ is the cross-sectional area of the resonator. Using $\lambda$ in Table IV and $J_{\text{mwa}}$ from the simulation, we obtain the following values of $L_{K1}$: 10 nH/m for Al-0L; 13 nH/m for Al-5L; and 2.5 nH/m for Al-10L. These results suggest that the difference in $f_0$ of Al-5L and Al-10L is due to the reduction of the kinetic inductance, i.e., the penetration depth, by thicker Al cladding. However, the difference in $f_0$ of Al-0L and Al-5L is not easy to understand. The Al-Nb interface might contribute to the inductance, but the mechanism is unclear.

Lastly, from Fig. 3, we found that 5 nm Al-cladding is a good choice for applications in X-band ESR of $g = 2$ electron spin systems, which require a magnetic field of about 0.35 T. However, if the film thickness or the film quality of the Nb layers is significantly different from that of Al-0L, then the optimal thickness of the Al layer may vary.

VI. CONCLUSION

In conclusion, we found that nonlinear responses of pure Nb microstrip resonators were induced by local Joule heating, while that of Al-clad resonators was induced by global heating. This qualitative change in non-
linear responses was likely due to Al current bypasses whose superconductivity is strengthened by the proximity effect between the Al and the Nb layers. This proximity effect was found to be controllable by tuning the Al layer thickness: as the thickness of the Al layer increases, $\lambda_0$, $H_c$, and $\rho_n$ decrease. Improving the film quality enhanced the microwave critical current density, but it did not result in a qualitative change in nonlinear responses. Thus, our study showed that Al cladding is an effective way to eliminate nonlinear responses induced by local Joule heating, resulting in improved microwave power handling capability.

Strong microwave power handling capability will allow us to control spins or solid-state qubits efficiently.\cite{2050-2223-12-9-097008} Hence, this work will be useful for magnetic resonance applications as well as quantum information processing.

SUPPLEMENTARY MATERIAL

See the supplementary material for details regarding solving the anisotropic GL equations (Sec. S1), extracting the loss parameters (Sec. S2), and magnetic hysteresis in a finite $H_\perp$ (Sec. S3), $S_21$ curves at various $P_{in}$ in a modest $H_\parallel$ are shown in Fig. S3.

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Engineering Nonlinear Response of Superconducting Niobium Microstrip Resonators via Aluminum Cladding: Supplementary Material

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S1 Anisotropic Ginzburg–Landau Equations

The anisotropic Ginzburg–Landau (GL) equations are given by (in SI units) \cite{1}

\[
\alpha \psi + \beta |\psi|^2 \psi + \frac{1}{2} \left( \frac{\hbar}{i} \nabla - e_s \vec{A} \right) \cdot \left[ \frac{1}{m^*} \right] \cdot \left( \frac{\hbar}{i} \nabla - e_s \vec{A} \right) \psi = 0, \tag{S1}
\]

\[
\frac{1}{\mu_0} \nabla \times \nabla \times \vec{A} = \frac{e_s \hbar}{2i} \left[ \frac{1}{m^*} \right] \cdot \left( \psi^* \nabla \psi - \psi \nabla \psi^* \right) - e_s^2 |\psi|^2 \left[ \frac{1}{m^*} \right] \cdot \vec{A}, \tag{S2}
\]

where \( \psi = \psi(x, y, z, t) \) is the complex order parameter; \( \alpha \) and \( \beta \) are phenomenological parameters; \( e_s \) is the charge of the superconducting electron; \( \vec{A} \) is the magnetic vector potential; \( \phi \) is the electric potential; and \([1/m^*]\) is the anisotropic effective mass tensor defined by

\[
\left[ \frac{1}{m^*} \right] = \begin{bmatrix}
1/m_{\parallel}^* & 0 & 0 \\
0 & 1/m_{\perp}^* & 0 \\
0 & 0 & 1/m_{\parallel}^*
\end{bmatrix}.
\]

In the anisotropic GL equations, this anisotropic effective mass is responsible for anisotropy in superconducting parameters. Here, we define the anisotropy parameter \( \gamma \) as

\[
\gamma = \sqrt{\frac{m_{\perp}^*}{m_{\parallel}^*}}.
\]

Then, we have the following relations:

\[
\frac{\xi_{\parallel}}{\xi_{\perp}} = \frac{\lambda_{\parallel}}{\lambda_{\perp}} = \frac{H_{c2}^\parallel}{H_{c2}^\perp} = \gamma.
\]

We transform the GL equations into dimensionless quantities by measuring length in units of the in-plane penetration depth \( \lambda_{\parallel} (\equiv \sqrt{m_{\parallel}^*/\mu_0 e_s^2 |\alpha|}) \); fields in units of \( \sqrt{2} H_c \), where \( H_c (\equiv \sqrt{\alpha^2/\mu_0 \beta}) \) is the thermodynamic critical field; and order parameter in units of \( \psi_0 (\equiv \sqrt{|\alpha|/\beta}) \). After the transformation, we introduced a time-dependent term from the time-dependent GL equations (Eqs. (S7) and (S8) of Ref. [2]) to imitate cooling procedures.
For simplicity, the time-dependent term is assumed isotropic. This can be justified by the argument that we are only interested in the steady-state solutions. Then Eqs. (S1) and (S2) are written as

\[
\frac{\partial \psi}{\partial t} = -\left( \frac{i}{\kappa} \nabla + \vec{A} \right) \cdot \left( \frac{i}{\Gamma^2} \nabla + \vec{A} \right) \psi + \psi - |\psi|^2 \psi, 
\]

\[
\sigma_n \frac{\partial \vec{A}}{\partial t} = \frac{1}{2i\kappa} \left[ \frac{1}{\Gamma^2} \right] \cdot (\psi^* \nabla \psi - \psi \nabla \psi^*) - |\psi|^2 \left[ \frac{1}{\Gamma^2} \right] \cdot \vec{A} - \nabla \times \nabla \times \vec{A},
\]

where \( \kappa = \lambda / \xi \) is the GL parameter; \( \sigma_n \) is the inverse of the residual resistivity; and \( 1/\Gamma^2 \) is a tensor given by

\[
\left[ \frac{1}{\Gamma^2} \right] = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1/\gamma^2 & 0 \\ 0 & 0 & 1 \end{bmatrix}.
\]

To solve Eqs. (S3) and (S4), we used COMSOL Multiphysics 5.1. The general form of partial differential equations in COMSOL Multiphysics is

\[
e_x \frac{\partial^2 \mathbf{u}}{\partial t^2} + d_a \frac{\partial \mathbf{u}}{\partial t} + \nabla \cdot \mathbf{\Gamma} = \mathbf{f}.
\]

All geometries were assumed to be two-dimensional systems on the \( xy \) plane. The applied magnetic field \( \vec{H}_a \) is assumed along the \( z \) direction. In this case, \( \mathbf{u} = (u_1, u_2, u_3, u_4, u_5)^\top \), where \( \top \) is the transpose. The variables are given by \( u_1(x, y, t) = \text{Re}(\psi(x, y, t)) \), \( u_2(x, y, t) = \text{Im}(\psi(x, y, t)) \), \( u_3(x, y, t) = A_x(x, y, t) \), and \( u_4(x, y, t) = A_y(x, y, t) \), respectively. An auxiliary variable \( u_5 \) is always zero. In Eq. (S5), \( e_a \) is a zero matrix. Others can be written as

\[
d_a = \begin{bmatrix} 1 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & \sigma_n & 0 & 0 \\ 0 & 0 & 0 & \sigma_n & 0 \\ 0 & 0 & 0 & 0 & 0 \end{bmatrix}, \quad \mathbf{\Gamma} = \begin{bmatrix} -\partial_x u_1/\kappa^2, -\partial_y u_1/(\gamma\kappa)^2 \\ -\partial_x u_2/\kappa^2, -\partial_y u_2/(\gamma\kappa)^2 \\ 0, \partial_x u_4 - \partial_y u_3 - \mu_0 H_a \\ -\partial_x u_4 + \partial_y u_3 + \mu_0 H_a, 0 \\ u_1, u_4 \end{bmatrix}^\top, \\
\mathbf{f} = \begin{bmatrix} (\partial_x u_3 + \partial_y u_4/\gamma^2) u_2/\kappa + 2(u_3 \partial_x u_2 + u_4 \partial_y u_2/\gamma^2)/\kappa - (u_3^2 + u_4^2) u_1 + u_1 - (u_1^2 + u_2^2) u_1 \\ -(\partial_x u_3 + \partial_y u_4/\gamma^2) u_1/\kappa - 2(u_3 \partial_x u_1 + u_4 \partial_y u_1/\gamma^2)/\kappa - (u_3^2 + u_4^2) u_2 + u_2 - (u_1^2 + u_2^2) u_2 \\ (u_1 \partial_x u_2 - \partial_y u_3) u_3 - (u_3^2 + u_4^2) u_3 \\ (u_1 \partial_y u_2 - \partial_x u_1) u_3/(\gamma^2 \kappa) - (u_3^2 + u_4^2) u_4/\gamma^2 \\ \partial_x u_4 + \partial_y u_4 + u_5 \end{bmatrix}.
\]

The boundary conditions were implemented using “zero flux” \(-\vec{n} \cdot \mathbf{\Gamma} = \mathbf{G}\), where \( \mathbf{G} = [0, 0, 0, 0]^\top \). The details of this implementation is described in Refs. [2, 3].

### S2 Extracting Loss Parameters from the Parallel Field Data

To extract loss parameters from the parallel field data (Fig. 4 in the main text), we should calculate the expected resonance frequency \( f \) and the quality factor \( Q \) as a function of external magnetic field \( H \). In the following, we introduce some formulas required for those calculations (see Sec. II of Ref. [2] for more information).

The magnetic field dependent parts of \( f \) and \( Q \) are given by

\[
\frac{f^{-2}(H) - f_0^{-2}}{f_0^{-2}} = \frac{L(H) - L_0}{L_0}, \quad \frac{1}{Q(H)} = \frac{P_{\text{diss}}(H)}{2\pi f(H)U_{\text{em}}(H)},
\]

where \( f_0 \) is the resonance frequency at zero-field; \( L_0 \) is the effective inductance per unit length (at zero-field); \( Q_0 \) is the quality factor at zero-field; \( P_{\text{diss}} \) is the dissipated power per unit length; and \( U_{\text{em}} \) is the stored electromagnetic energy per unit length. Since \( L \) is defined by the relation \( U_{\text{em}} = L|I|^2/2 \), where \( I \) is the total microwave current, the quantities we need to calculate are \( U_{\text{em}} \) and \( P_{\text{diss}} \).
Consider a microstrip line oriented along the z axis with its width along the x axis and thickness along the y axis. In this configuration, $U_{em}$ is defined by

$$U_{em}(H) = \frac{1}{2} \int_{all} \mu_0 |H_{mw}(x, y, \lambda(H))|^2 dxdy + \frac{1}{2} \int_{sc} \frac{\rho_2(x, y, H)}{\omega} |J_{mw}(x, y, \lambda(H))|^2 dxdy \quad (S7)$$

where $\mu_0$ is the vacuum permeability; $\rho$ is the complex resistivity $\rho_1 + i\rho_2$; $H_{mw}$ is the microwave magnetic field strength; $J_{mw}$ is the microwave current density; $\lambda$ is the penetration depth; $\omega/2\pi$ is the frequency of an applied electromagnetic field; and “sc” stands for “inside superconducting media”. Next, $P_{diss}$ is defined by

$$P_{diss}(H) = \frac{1}{2} \int_{sc} \rho_1(x, y, H) |J_{mw}(x, y, \lambda(H))|^2 dxdy. \quad (S8)$$

In Eqs. (S7) and (S8), $J_{mw}$ and $H_{mw}$ can be simulated by the Maxwell equations and the London equations (see Sec. S2 of Ref. [2] for details of the simulation). For the complex resistivity, we employ the two-fluid model. The complex conductivity based on the two-fluid model $\sigma_{tf,1} = i\sigma_{tf,2}$ is given by [4]

$$\sigma_{tf,1} = \frac{n_s}{n_{tot}} \sigma_n, \quad \sigma_{tf,2} = \frac{n_s e_s^2}{m_s \omega} = \frac{1}{\omega \mu_0 \lambda^2}; \quad (S9)$$

where $n_s$ is the local number density of superconducting electrons; $n_{tot}$ is the local number density of normal electrons; $n_{tot}$ is the total number density of conduction electrons; $\sigma_n$ is the inverse of the residual resistivity $\rho_n$; $e_s$ is the charge of a superconducting electron; and $m_s$ is the mass of a superconducting electron. The corresponding complex resistivity $\rho_{tf,i}$ is given by $\rho_{tf,i} = \sigma_{tf,i}/(\sigma_{tf,1}^2 + \sigma_{tf,2}^2)$.

Once we solve the anisotropic GL equations as described in Sec. S1, $n_s$ can be calculated using the relation $n_s(x, y, H) = |\psi(x, y, H)|^2$. As the GL theory does not give $n_n$, we introduce an empirical expression for $n_n$ with an additional exponent $\beta$:

$$\frac{n_n(H)}{n_{tot}} = \left[1 - \frac{n_n(H)}{n_n(0)}\right]^\beta. \quad (S10)$$

The procedure for extracting the loss parameters used in this work is identical to that in Ref. [2], except two parameters, $\lambda_0$ and $\kappa$ in Ref. [2], become $\lambda_0^\parallel$ and $\gamma \kappa^\parallel$ due to the anisotropy. A brief description of the procedure is as follows (see Sec. S3 of Ref. [2] for more details):

1. Calculate $n_s$ as a function of $H^\parallel$ by solving the GL equations; $\lambda_0^\parallel$ and $\gamma \kappa^\parallel$ are required for this step.
2. Since $n_s \propto \lambda^{-2}$ [Eq. (S9)], $\lambda^\parallel(H^\parallel)$ is obtained by $(\lambda^\parallel_0/\lambda^\parallel)^2 = n_s(H^\parallel)/n_s(0)$. Once $\lambda^\parallel$ is known, $J_{mw}$ and $H_{mw}$ can be simulated. Here, note that $\lambda^\parallel$ is responsible for those simulations because the microwave current flows parallel to the film.
3. $f^{-2}$ is reconstructed theoretically using Eqs. (S6), (S7), and (S9); $H_c$ is required for this step.
4. Repeat steps 1–3 until the theoretical $f^{-2}$ is sufficiently close to the experimental results. Then, $\lambda_0^\parallel$, $\gamma \kappa^\parallel$, and $H_c$ are determined. Here, $\lambda_0^\parallel$ is determined by the slope of $f^{-2}(H^\parallel)$ below $H^\parallel_c$ because the slope of $f^{-2}(H^\parallel)$ was determined primarily by geometrical constriction, given by $d/\lambda_0^\parallel$, where $d$ is the thickness of the whole trilayer; $\gamma \kappa^\parallel$ is chiefly determined by $H^\parallel_c$.
5. Using $n_s(H^\parallel)$ from the $f^{-2}$ data, $Q_0$, $\beta$, and $\rho_n$ are determined by the $Q^{-1}$ data using a similar procedure and Eqs. (S6)–(S10).

During the calculation, as in Ref. [2], the ground plane’s contribution to the resonator properties was assumed to be negligible because this contribution to the microwave current density is just a few percent. The same assumption was also applied for the loss parameters associated with vortex motion.
**S3 Magnetic Hysteresis in a Finite Perpendicular Field**

As mentioned in Sec. IV, we found that applying a high microwave current in a magnetic field perpendicular to the film $H_\perp$ results in magnetic hysteresis. To explore the effect of $H_\perp$ on the nonlinear behavior, which will be crucial for resonators misaligned with the field, we measured the microwave power dependence of Al-5L with a finite $H_\perp$.

$H_\perp$ was applied by tilting the resonator in a background magnetic field parallel to the microwave current $H_{ bg}$ using the goniometer mentioned in Sec. II. $H_\perp$ is obtained by $H_\perp = H_{ bg} \sin \theta$, where $\theta$ is the tilt angle. In this work, $\mu_0 H_{ bg} = 0.35 \text{T}$. Two different types of cooling procedure were used: zero-field cooling (ZFC) and heat pulsing (HP). For the ZFC procedure, the resonator is cooled without any magnetic field. For the HP procedure, a heat pulse is applied to completely suppress superconductivity, then the resonator is cooled back in field to the target temperature. The HP procedure is used to ensure a uniform vortex distribution and suppress the Meissner current as much as possible such that vortex motion becomes the dominant loss mechanism [2].

After applying a high microwave current, significant changes in $f$ and $Q$ were observed as shown in Fig. S1. These changes at various $H_\perp$ are presented in Fig. S2(a). The data were taken by the following procedure: First, $H_\perp$ is applied after ZFC by tilting the resonator. Then the incident power on the input capacitor of the resonator $P_{ in}$ is applied from low power ($-57 \text{ dBm}$) to the power called the current annealing power $P_{ CA}$. Note that as $P_{ in}$ increases, the spectrum not only shows the Duffing-like nonlinearity but also shifts to a higher frequency as shown in Fig. S1 (solid lines). Once $P_{ in}$ reaches $P_{ CA} (-7 \text{ dBm} \text{ for Fig. S1})$, $P_{ in}$ is reduced to $-57 \text{ dBm}$. We call the procedure up to this point the initial current annealing. After the initial current annealing, no spectrum shift is observed for $P_{ in} \leq P_{ CA}$ (dashed lines). The position of the spectrum from a low-power measurement is completely determined by the highest power prior to the low-power measurement regardless of a history of $P_{ in}$.

To understand the origin of this hysteresis induced by high microwave current, we first consider the $H_\perp$ dependence of $f^{-2}$ and $Q$ without the current annealing [Fig. S2(a)]. After the HP procedure, $f^{-2}$ and $Q^{-1}$ vary linearly with the field, i.e., $Q$ is roughly proportional to the inverse of $H_\perp$, indicating continuous occupation of vortices [2]. Hence, the magnetic field dependence after the HP procedure is governed by vortex motion. A key feature of the data after the ZFC procedure without current annealing is that an anomaly (peak/dip) appears at the field $B$ (6.7 mT) in the $f^{-2}$ data. As shown in our previous work [2], this frequency anomaly is an indication of complete suppression of the Bean–Livingston type edge barrier [5]; hence, the field $B$ is the vortex penetration field perpendicular to the film $H_p$. Below this field, the magnetic field dependences of the microwave properties are governed by quasiparticles generated by the Meissner current; above this field, vortex motion is the dominant mechanism. If a superconducting resonator is in a metastable state due to the edge barrier, the high microwave current can change the resonator state via suppressing the edge barrier [6], resulting in different $f$ and $Q$ values. This suppression of the edge barrier by the microwave current is indeed indicated by the result in Fig. S2(a) that the frequency anomaly becomes weaker as $P_{ CA}$ increases.

To reveal the physical processes behind the magnetic hysteresis, we use a plot of $Q$ vs. $f^{-2}$ [Fig. S2(b)]. The motivation of this plot is to represent the characteristic relation between the real and imaginary parts of the complex resistivity for each contribution—either quasiparticle generation or vortex motion [2]. In this plot, a process of varying quasiparticle numbers evolves horizontally keeping $Q^{-1}$ constant (an arrow labeled “qp”), while a process of varying vortex numbers evolves as a nearly vertical line (an arrow labeled “vm”). The reason is that, for our device in the range of magnetic fields studied, quasiparticle generation is a very inductive process such that the kinetic energy of existing vortices by the microwave current, or a “shaking” of the vortices. The displacement of vortices during the shaking expels the Meissner current.

In conclusion, we analyzed magnetic hysteresis in $f$ and $Q$ induced by a high microwave current when $H_\perp$ was higher than a certain level. We revealed the physical processes behind this using a plot of $Q$ vs. $f^{-2}$. By doing this, we found that the observed hysteresis was induced by suppression of the edge barrier and consequent vortex injection.
Figure S1: $S_{21}$ curve shift of Al-5L due to a strong microwave current at $\mu_0H_\perp = 6.7$ mT. Annotated powers ($P_n$) were applied sequentially. $f_\perp$ is the resonance frequency before the initial current annealing. Solid lines are the results from the initial current annealing and dashed lines are from the second annealing. Results from further sequences are identical to the second one. The sweep direction was from low to high frequency. The meaning of the annotation “Field B” can be found in Fig. S2(a). The measurement temperature was about 0.2 K.

Figure S2: (a) Final $f^{-2}$ and $Q$ for Al-5L measured with $P_m = -57$ dBm after the initial current annealing. (b) A $Q$ vs. $f^{-2}$ plot of the data in (a). Arrows labeled A, B, and C indicate the direction of evolution by the current annealing. In (a,b), symbols with different colors mean that the data were taken with a different $P_{CA}$, while small black circles were taken without current annealing; small empty circles were taken after the HP procedure. All lines are guides to the eye. (c) The Meissner current and vortices configuration before and after the current annealing at the designated fields. The gray gradient indicates the schematic distribution of the Meissner current density; the darker area is higher current density. The dark gray circles are vortices. The measurement temperature was about 0.2 K.
Figure S3: $S_{21}$ resonance curves at various $P_{in}$ in a modest $H_\parallel$. The sweep direction was from low to high frequency. The measurement temperature was about 0.2 K.

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