
Higher-order harmonics in Josephson tunnel junctions due to series inductance

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S1. EXPERIMENTAL SETUP

Figure S1 represents the experimental wiring. All experiments were conducted in a Leiden CF-450 dilution refrigerator operating at a base temperature of approximately 25 mK. The flux bias for the five SQUIDS is provided by a superconducting coil mounted to the lid of the sample package. The equipment used in this experiment is given in Table SI.

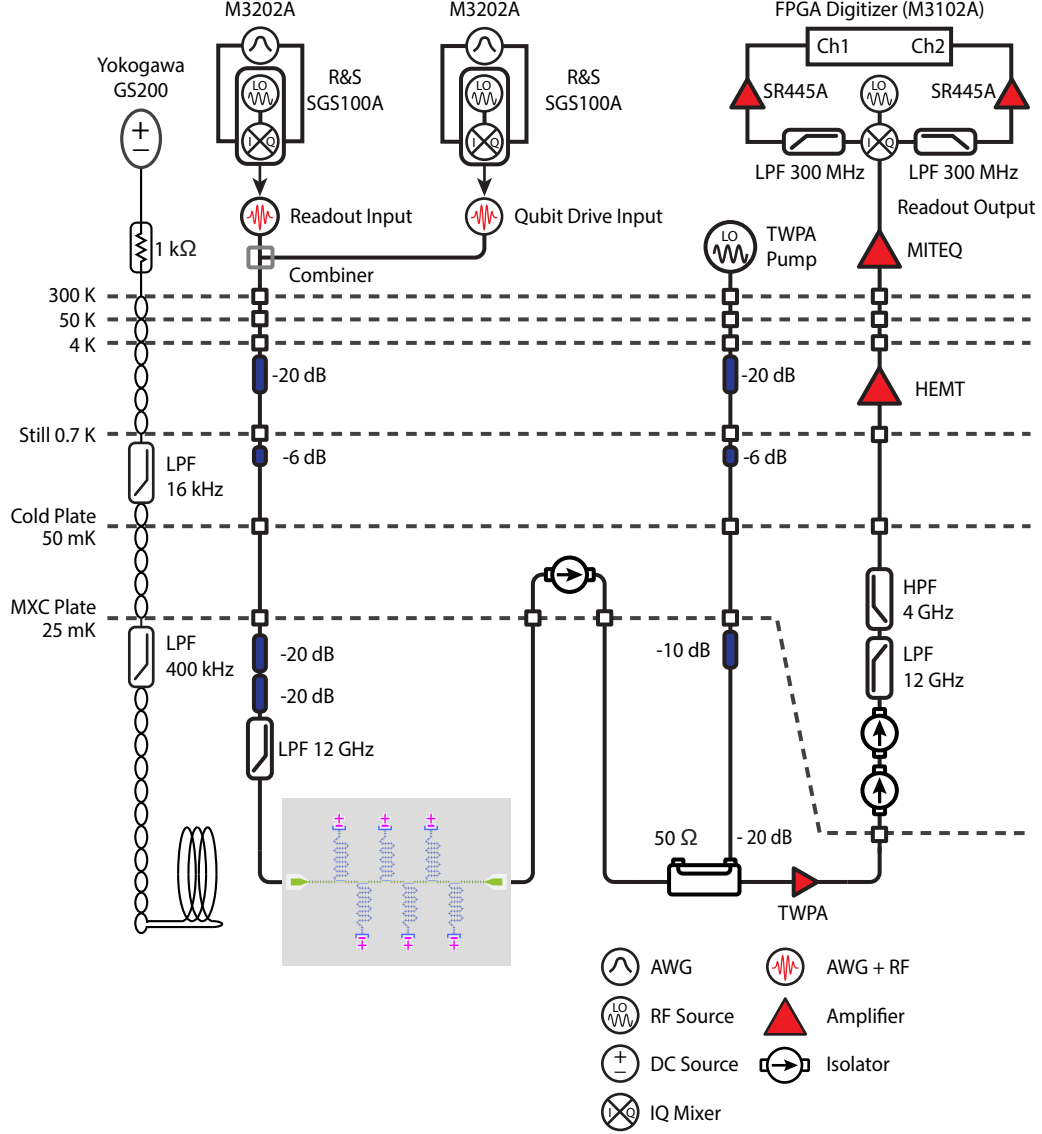


FIG. S1. Wiring diagram of the experimental setup.

TABLE SI. Summary of control equipment.

Component	Manufacturer	Model
Dilution Refrigerator	Leiden Cryogenics	CF-450
RF Source	Rohde & Schwarz	SGS100A
DC Source	Yokogawa	GS200
Control Chassis	Keysight	M9019A
Arbitrary Waveform Generator (AWG)	Keysight	M3202A
Digitizer	Keysight	M3102A

S2. POTENTIAL IN THE PRESENCE OF ANDREEV HARMONICS AND SERIES INDUCTANCE

A. Andreev process

The potential energy of a Josephson tunnel junction described by the Andreev process is given by

$$U(\varphi_J) = -\Delta \sum_{n=1}^N \sqrt{1 - T_n \sin^2 \left(\frac{\varphi_J}{2} \right)}, \quad (\text{S1})$$

where T_n is the transparency of the n^{th} conduction channel, and N is the number of the channels. φ_J is the phase across the junction. For a superconductor-insulator-superconductor (SIS) tunnel junction, these two intrinsic parameters are estimated to be $N \sim 10^6$, and $T_n \sim 10^{-6}$ [1]. Assuming T_n is small, we can expand the square root to obtain

$$U(\varphi_J) = -\Delta \sum_{n=1}^N \left(1 - \frac{1}{2} T_n \sin^2 \left(\frac{\varphi_J}{2} \right) - \frac{1}{8} T_n^2 \sin^4 \left(\frac{\varphi_J}{2} \right) - \frac{1}{16} T_n^3 \sin^6 \left(\frac{\varphi_J}{2} \right) - \dots \right). \quad (\text{S2})$$

Using trigonometric identities, the potential can be expressed as a sum of harmonics

$$U(\varphi_J) = -E_{J1,A} \cos(\varphi_J) + E_{J2,A} \cos(2\varphi_J) - E_{J3,A} \cos(3\varphi_J) + \dots, \quad (\text{S3})$$

where the coefficients satisfy

$$E_{J1,A} = \Delta \sum_{n=1}^N \left(\frac{1}{4} T_n + \frac{1}{16} T_n^2 + \frac{15}{512} T_n^3 + \dots \right) \quad (\text{S4})$$

$$E_{J2,A} = \Delta \sum_{n=1}^N \left(\frac{1}{64} T_n^2 + \frac{3}{256} T_n^3 + \frac{35}{4096} T_n^4 - \dots \right) \quad (\text{S5})$$

$$E_{J3,A} = \Delta \sum_{n=1}^N \left(\frac{1}{512} T_n^3 + \frac{5}{2048} T_n^4 + \frac{315}{131072} T_n^5 + \dots \right). \quad (\text{S6})$$

Taking only the leading order term for each E_{J1} and E_{J2} , the coefficients for the intrinsic Andreev process can be approximated as

$$E_{J1,A} \approx \Delta \sum_{n=1}^N \frac{1}{4} T_n \quad (\text{S7})$$

$$E_{J2,A} \approx \Delta \sum_{n=1}^N \frac{1}{64} T_n^2 \quad (\text{S8})$$

Assuming a large number of channels $N \gg 1$ and that the transparency follows a distribution $T \sim \rho(T)$, the summation over all conduction channels can be approximated as an integral over the distribution [1].

$$\sum_{n=1}^N f(T_n) \approx N \int_0^1 \rho(T) f(T) dT. \quad (\text{S9})$$

The ratio between the first and the second harmonics becomes

$$\frac{E_{J2,A}}{E_{J1,A}} = \frac{\frac{1}{64} \sum_{n=1}^N T_n^2}{\frac{1}{4} \sum_{n=1}^N T_n} \approx \frac{\frac{N}{64} \int_0^1 T^2 \rho(T) dT}{\frac{N}{4} \int_0^1 T \rho(T) dT} = \frac{1}{16} \frac{\int_0^1 T^2 \rho(T) dT}{\int_0^1 T \rho(T) dT}, \quad (\text{S10})$$

which only depends on the distribution of the transparency. In this study, we assume our junction size is large enough to provide a sufficiently large number of channels, so that the summation of the transparency (squared) is well approximated by the integral. Moreover, we assume the same transparency distribution $\rho(T)$ for all devices measured, so that varying the junction area only affects the number of channels N , which does not appear in the final ratio. This assumption is reasonable for our devices because they were fabricated in the same deposition/oxidation run. Under these assumptions, the ratio of the second harmonic

$$\frac{E_{J2,A}}{E_{J1,A}} \approx \frac{1}{16} \frac{\int_0^1 T^2 \rho(T) dT}{\int_0^1 T \rho(T) dT} = \gamma \quad (\text{S11})$$

remains constant across the devices under test.

B. Full-circuit model vs second-harmonic approximation

In this section, we discuss the validity of the harmonic approximation used to model the effect of the inductance in series with the junction. We compare the harmonic approximation of the SQUID potentials to a full-circuit analysis, and compute the difference between the energy eigenvalues of the two models for the validity of the harmonic approximation. We find that while the harmonic approximation is valid for our circuit parameters, it is not necessarily valid for more typical transmon parameters and therefore should be used with caution.

We first derive the harmonic approximation used in the main text, following Ref. [1]. Figure S2 shows a Josephson junction in series with an inductor, where the junction capacitance has temporarily been neglected. In addition, in this section we will assume that there are no intrinsic higher harmonics.

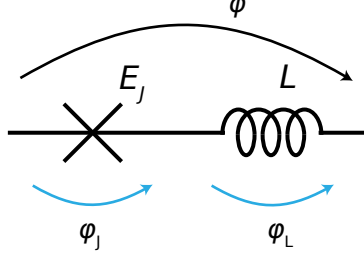


FIG. S2. A Josephson junction in series with a linear inductor. The junction capacitance is temporarily neglected.

The branch phase variables for the junction and the inductor are denoted as φ_J and φ_L , respectively, and $\varphi = \varphi_J + \varphi_L$ represents the total phase across the series. The potential of this circuit is given by

$$U(\varphi, \varphi_J) = -E_J \cos(\varphi_J) + \frac{1}{2L} \left(\frac{\Phi_0}{2\pi} \right)^2 (\varphi - \varphi_J)^2. \quad (\text{S12})$$

To eliminate φ_J from the potential, we make the approximation that the current through the junction is equal to the current through the inductor:

$$I_c \sin \varphi_J = \frac{1}{L} \left(\frac{\Phi_0}{2\pi} \right) (\varphi - \varphi_J), \quad (\text{S13})$$

which is equivalent to

$$E_J \sin \varphi_J = E_L (\varphi - \varphi_J), \quad (\text{S14})$$

where $E_L = \frac{1}{L} \left(\frac{\Phi_0}{2\pi} \right)^2$ is the inductive energy. As discussed in more detail below, we stress that this approximation neglects the junction capacitance and is therefore only valid for certain circuit parameter regimes and should be carefully checked. Under this approximation, the φ_J degree of freedom can be eliminated from the potential by substituting Eq. (S14) into Eq. (S12).

To this end, we first present a simple method of successive approximation to calculate the leading-order second-harmonic coefficient. Rewriting Eq. (S14), we obtain

$$\varphi - \varphi_J = \frac{E_J}{E_L} \sin \varphi_J. \quad (\text{S15})$$

Assuming $E_J \ll E_L$, the zeroth-order equation for φ_J is given by

$$\varphi - \varphi_J^{(0)} = 0. \quad (\text{S16})$$

Consequently, we find the first-order equation as

$$\varphi - \varphi_J^{(1)} = \frac{E_J}{E_L} \sin \varphi_J^{(0)} = \frac{E_J}{E_L} \sin \varphi. \quad (\text{S17})$$

Substituting $\varphi_J^{(1)}$ into Eq. (S12) yields

$$\begin{aligned} U(\varphi) &= -E_J \cos \left(\varphi - \frac{E_J}{E_L} \sin \varphi \right) + \frac{1}{2} E_L \left(\frac{E_J}{E_L} \sin \varphi \right)^2 \\ &\approx -E_J \left[\cos \varphi + \frac{E_J}{E_L} \sin^2 \varphi \right] + \frac{1}{2} E_L \left(\frac{E_J}{E_L} \sin \varphi \right)^2 \\ &= -E_J \cos \varphi - \frac{1}{2} \frac{E_J^2}{E_L} \sin^2 \varphi \end{aligned} \quad (\text{S18})$$

Using the double-angle formula and dropping the constant term, we obtain

$$U(\varphi) \approx -E_J \cos \varphi + \frac{E_J^2}{4E_L} \cos(2\varphi). \quad (\text{S19})$$

We also derive the above result following the more detailed method discussed in Ref. [1]. The generalized harmonic expansion is given by

$$U(\varphi) = - \sum_{n \geq 1} E_{Jn,L} \cos(n\varphi), \quad (\text{S20})$$

where the coefficients $E_{Jn,L}$ satisfy

$$\begin{aligned} E_{J1,L} &= E_J \left[1 - \frac{1}{8} \left(\frac{E_J}{E_L} \right)^2 + \frac{1}{192} \left(\frac{E_J}{E_L} \right)^4 + \dots \right] \\ E_{J2,L} &= E_J \left[-\frac{1}{4} \left(\frac{E_J}{E_L} \right) + \frac{1}{12} \left(\frac{E_J}{E_L} \right)^3 - \frac{1}{96} \left(\frac{E_J}{E_L} \right)^5 + \dots \right] \\ E_{J3,L} &= E_J \left[\frac{1}{8} \left(\frac{E_J}{E_L} \right)^2 - \frac{9}{128} \left(\frac{E_J}{E_L} \right)^4 + \frac{1}{64} \left(\frac{E_J}{E_L} \right)^6 + \dots \right] \\ E_{J4,L} &= E_J \left[-\frac{1}{12} \left(\frac{E_J}{E_L} \right)^3 + \frac{1}{15} \left(\frac{E_J}{E_L} \right)^5 - \frac{101}{4608} \left(\frac{E_J}{E_L} \right)^7 + \dots \right]. \end{aligned} \quad (\text{S21})$$

Truncating the summation at the second harmonic ($n = 2$) and retaining only the leading order terms, the potential is approximated as

$$U(\varphi) \approx -E_J \cos(\varphi) + \frac{E_J^2}{4E_L} \cos(2\varphi), \quad (\text{S22})$$

which is the same as Eq. (S19). By applying Eq. (S22) to each arm of the SQUID, which comprises a series connection of a junction and an inductor, the Hamiltonian of the circuit is approximately given by

$$\begin{aligned} H_{\text{SQUID}}(\phi_{\text{ext}}) &= 4E_C(\hat{n} - n_g)^2 - E_J^{(L)} \cos(\hat{\varphi}) - E_J^{(R)} \cos(\hat{\varphi} - \phi_{\text{ext}}) \\ &\quad + \frac{\left(E_J^{(L)}\right)^2}{4E_L} \cos(2\hat{\varphi}) + \frac{\left(E_J^{(R)}\right)^2}{4E_L} \cos(2(\hat{\varphi} - \phi_{\text{ext}})), \end{aligned} \quad (\text{S23})$$

where $E_C = e^2/2(C_J^{(L)} + C_J^{(R)} + C_{\text{sh}})$ includes the shunt and junction capacitances.

We now proceed with a full-circuit analysis to check the validity of this second-harmonic model. While above we set the junction current equal to the inductor current in order to eliminate the junction phases from the potential energy, here we will keep these phases and solve the Hamiltonian accounting for all three degrees of freedom [Fig. S3]. The full-circuit Hamiltonian is derived from the following Lagrangian

$$\begin{aligned} T &= \frac{1}{2} C_J^{(L)} \left(\frac{\Phi_0}{2\pi} \right)^2 \dot{\varphi}_1^2 + \frac{1}{2} C_J^{(R)} \left(\frac{\Phi_0}{2\pi} \right)^2 \dot{\varphi}_2^2 + \frac{1}{2} C_{\text{sh}} \left(\frac{\Phi_0}{2\pi} \right)^2 (\dot{\phi}_1 + \dot{\phi}_2)^2 \\ U &= \frac{1}{2} E_L (\phi_1^2 + \phi_2^2) - E_J^{(L)} \cos(\varphi_1) - E_J^{(R)} \cos(\varphi_2) \\ L_{\text{full}} &= T - U. \end{aligned} \quad (\text{S24})$$

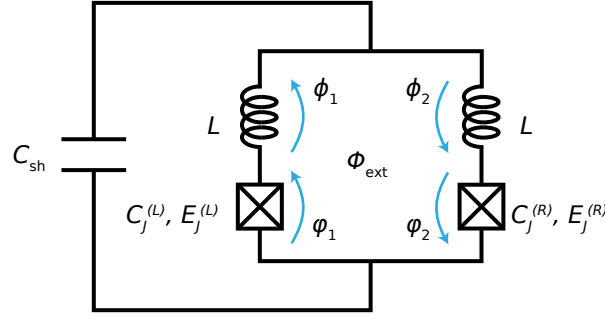


FIG. S3. A full-circuit model of the capacitively shunted SQUID including two series inductors.

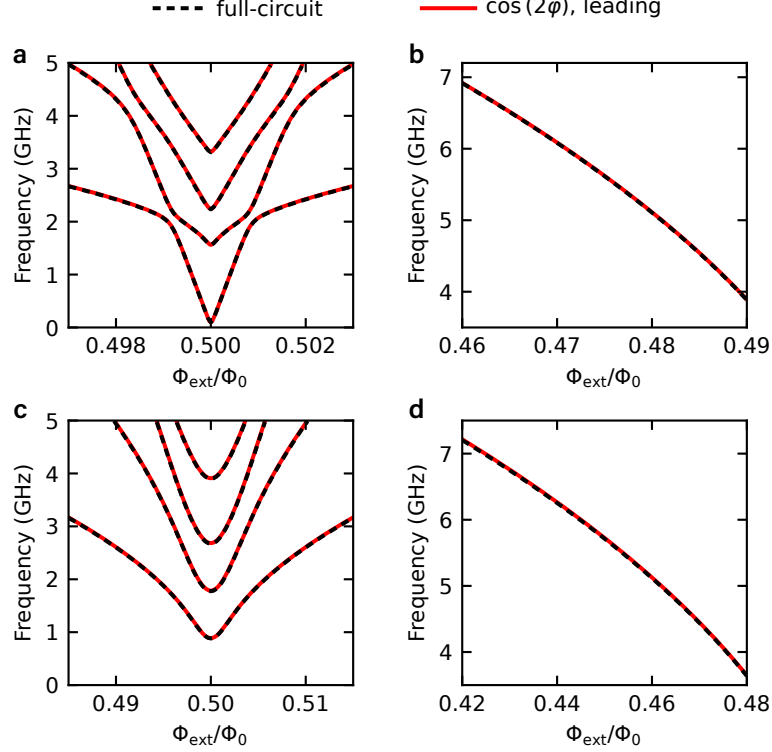


FIG. S4. Transition spectra of the full-circuit model (black dashed) and second-harmonic model (red solid). (a) and (b) show the spectra of the 6 μm device with $E_J \approx 230$ GHz, and (c) and (d) of the 2 μm device with $E_J \approx 80$ GHz.

By changing the variables as follows [2]

$$\begin{aligned}
 \varphi_1 &= \frac{\phi}{2} + \varphi \\
 \varphi_2 &= \frac{\phi}{2} - \varphi \\
 \phi_1 &= \theta - \frac{1}{2}\phi + \frac{\phi_{\text{ext}}}{2} \\
 \phi_2 &= -\theta - \frac{1}{2}\phi + \frac{\phi_{\text{ext}}}{2},
 \end{aligned} \tag{S25}$$

we reach the full-circuit Hamiltonian

$$\begin{aligned}
 H_{\text{full}} &= 4E_{C\theta} \hat{n}_\theta^2 + 4E_{C\phi} \hat{n}_\phi^2 + 4E_{C\varphi} (\hat{n}_\varphi - n_g)^2 - J\hat{n}_\theta(\hat{n}_\varphi - n_g) \\
 &\quad + E_L \hat{\theta}^2 + \frac{1}{4}E_L(\hat{\phi} - \phi_{\text{ext}})^2 - E_J^{(R)} \cos\left(\frac{\hat{\phi}}{2} - \hat{\varphi}\right) - E_J^{(L)} \cos\left(\frac{\hat{\phi}}{2} + \hat{\varphi}\right),
 \end{aligned} \tag{S26}$$

where

$$E_{C\theta} = \frac{e^2}{2} \left(\frac{1}{C_{\text{sh}}} + \frac{1}{2C_J} \right), \quad E_{C\phi} = \frac{e^2}{C_J}, \quad E_{C\varphi} = \frac{e^2}{4C_J}, \quad J = \frac{2e^2}{C_J}, \quad (\text{S27})$$

when $C_J^{(R)} \approx C_J^{(L)} = C_J$. Figure S4 shows the spectra of the full-circuit model (black dashed), which are obtained by directly diagonalizing Eq. (S26), and the spectra of the second-harmonic model (red solid) from Eq. (S23). We observe that, within our circuit parameter regime, the eigenenergies of the approximate Hamiltonian accurately match those of the full-circuit Hamiltonian. We also simulate the transition spectrum of a more general circuit, which includes an additional inductance outside the SQUID loop, connecting the SQUID and the shunt capacitor. From a conservative choice of the additional inductance (≈ 22 pH), we find the maximum energy difference between this model and Eq. S26 is less than the linewidth of the measured transition spectrum.

Moreover, we notice that the current-conservation-based harmonics approximation [Eq. (S20)] actually requires the junction capacitance to be sufficiently large. This becomes clearer when the series inductance is large. For example, with the transmon parameter regime used in Ref. [1], we observe a discrepancy between the second-harmonic approximation and the full-circuit analysis, as shown in Fig. S5. In this comparison [Fig. S5b], we also included the result of the Hamiltonian truncated at the fourth harmonic with higher powers of E_J/E_L in Eq. (S20) to check possible further corrections. Here, the Hamiltonian of the full-circuit [Fig. S5a] is derived from the Lagrangian

$$L_{\text{full}} = \frac{1}{2} C \dot{\Phi}^2 + \frac{1}{2} C_J (\dot{\Phi} - \dot{\Phi}_L)^2 + E_J \cos(\varphi - \varphi_L) - \frac{1}{2} E_L \varphi_L^2, \quad (\text{S28})$$

which leads to

$$H_{\text{full}} = 4(E_C + E_{CJ}) \hat{n}_L^2 + \frac{1}{2} E_L \hat{\varphi}_L^2 + 4E_C (\hat{n} - n_g)^2 - E_J \cos(\hat{\varphi} - \hat{\varphi}_L) + 8E_C (\hat{n} - n_g) \hat{n}_L, \quad (\text{S29})$$

where $E_C = e^2/2C$, $E_{CJ} = e^2/2C_J$. Here, $\varphi = 2\pi\Phi/\Phi_0$ and $\varphi_L = 2\pi\Phi_L/\Phi_0$ are the reduced flux variables across the shunt capacitor and the inductor, respectively. As outlined in the main text, the difference arises as the fluctuation of φ_L increases in small C_J regime.

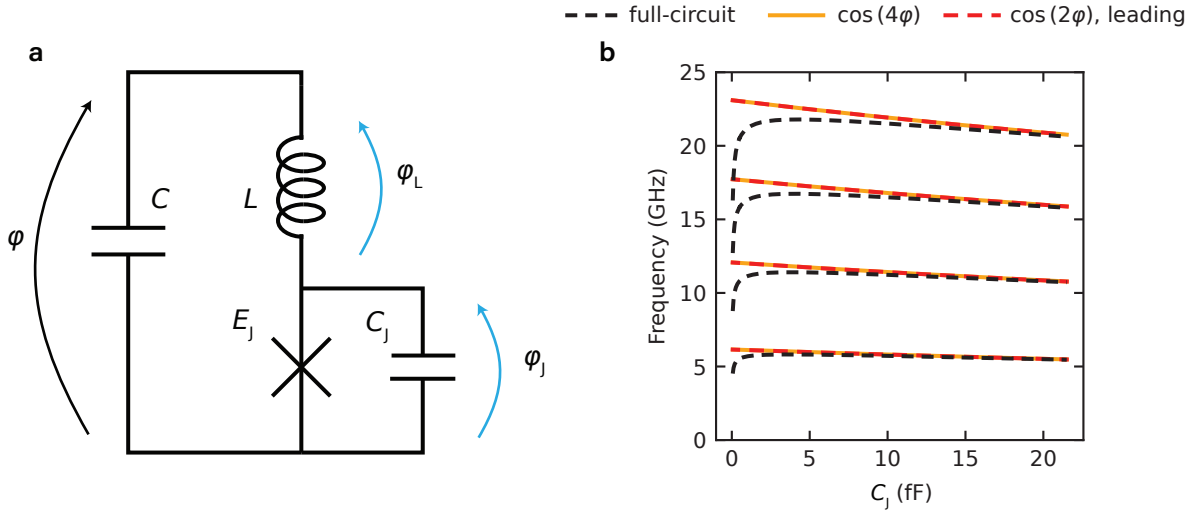


FIG. S5. (a) A full-circuit model of a transmon with a series inductance. (b) Differing energies of the second-harmonic approximation in low C_J regime (plot range: $C_J > 0.05$ fF). The fourth-harmonic approximation result (orange solid) overlaps with the second-harmonic approximation (red dashed). Circuit parameters are adopted from Ref. [1]: $C = 79$ fF, $E_J = 21.8$ GHz, $L = 380$ pH.

Keeping the discrepancy of the harmonics approximation for the inductance-induced correction in mind, we briefly re-analyze the fixed-frequency transmon spectra presented in Ref. [1] (three cooldown data for the KIT sample). We use two models: 1) the inductance-only model illustrated in Fig. S5a assuming no intrinsic Andreev harmonics, and

2) the same model as 1) but including the second Andreev harmonic $E_{J2,A}$. For the first model, we optimize L , C_J , and the resonator coupling strength J as global fit parameters across the three cooldown datasets. For the second model, we fix $J = 85$ MHz and optimize the shunt capacitance C as a global fit parameter to reduce the number of fit variables. Table. **SII- SIII** show possible sets of circuit parameters for the two models that agree with the data within 2 MHz. As such, it is possible that intrinsic harmonics were at play in the measured devices. However, the large uncertainty (in some cases by more than an order of magnitude) indicates that more data points are needed to obtain statistically reliable parameters. For example, this could be done using the SQUID structures as proposed in this work.

TABLE SII. Possible circuit parameter set for transmon spectra presented in Ref. [1] (KIT) using inductance-only model. Three cooldown data is combined.

parameter	E_{J1}/h (cd1) (GHz)	E_{J1}/h (cd2) (GHz)	E_{J1}/h (cd3) (GHz)	C (cd1) (fF)	C (cd2) (fF)	C (cd3) (fF)	C_J (fF)	L (pH)	J (MHz)
value	25.7	24.1	15.3	79.2	78.3	77.6	1.43	644	86.8
uncertainty	51.1	48.0	30.5	34.6	34.5	34.2	33.8	61.4	4.13

TABLE SIII. Possible circuit parameter set for transmon spectra presented in Ref. [1] (KIT) using inductance + second Andreev harmonic model. Three cooldown data is combined.

parameter	E_{J1}/h (cd1) (GHz)	E_{J1}/h (cd2) (GHz)	E_{J1}/h (cd3) (GHz)	E_{J2}/h (cd1) (GHz)	E_{J2}/h (cd2) (GHz)	E_{J2}/h (cd3) (GHz)	C (fF)	L (pH)	C_J (fF)
value	24.8	23.5	15.0	0.277	0.193	0.0367	78.8	406	1.41
uncertainty	43.9	41.4	26.5	1.75	1.22	0.251	36.9	706	36.5

C. Potential in the presence of both sources

In this section we derive the equation for the total effective second harmonic $E_{J2} \approx E_{J2,A} + E_{J2,L}$ in the presence of both Andreev process and series inductance. Here, we adopt the method of perturbatively optimizing φ_L to minimize the potential discussed in Refs. [3, 4], which produces the same result as the method used to derive Eq. (S22).

In the presence of the intrinsic Andreev second harmonic, $E_{J2,A} = \gamma E_{J1}$, the potential energy of the series in Fig. S2 is given by

$$\begin{aligned} U &= -E_{J1} \left[\cos(\varphi - \varphi_L) - \gamma \cos(2\varphi - 2\varphi_L) \right] + \frac{1}{2} E_L \varphi_L^2 \\ &= -E_{J1} \left[\cos(\varphi) \cos(\varphi_L) + \sin(\varphi) \sin(\varphi_L) \right] \\ &\quad + \gamma E_{J1} \left[\cos(2\varphi) \cos(2\varphi_L) + \sin(2\varphi) \sin(2\varphi_L) \right] + \frac{1}{2} E_L \varphi_L^2, \end{aligned} \quad (\text{S30})$$

where $E_{J1} = E_{J1,A}$ is the fundamental harmonic of the Andreev process. Expanding the cosines and sines up to the second order of φ_L , we obtain

$$\begin{aligned} U &= -E_{J1} \left[\cos(\varphi) \left(1 - \frac{1}{2} \varphi_L^2 \right) + \sin(\varphi) \varphi_L \right] \\ &\quad + \gamma E_{J1} \left[\cos(2\varphi) (1 - 2\varphi_L^2) + \sin(2\varphi) (2\varphi_L) \right] + \frac{1}{2} E_L \varphi_L^2, \end{aligned} \quad (\text{S31})$$

which is minimized when

$$\varphi_L = \frac{E_{J1} \sin(\varphi) - 2\gamma E_{J1} \sin(2\varphi)}{E_L + E_{J1} \cos \varphi - 4\gamma E_{J1} \cos(2\varphi)}. \quad (\text{S32})$$

Again, we expand the right-hand side of the equation using $1/(1-x) \approx 1+x+x^2+\dots$, where the expansion variable x in this case is

$$x = \frac{E_{J1}}{E_L} \left(-\cos(\varphi) + 4\gamma \cos(2\varphi) \right). \quad (\text{S33})$$

Substituting Eq. (S32) and Eq. (S33) into Eq. (S31), we obtain the following harmonic expansion.

$$U = - \sum_{n \geq 1} E_{Jn,\text{eff}} \cos(n\varphi), \quad (\text{S34})$$

where

$$\begin{aligned} E_{J1,\text{eff}} &= E_{J1} \left[1 - \gamma \left(\frac{E_{J1}}{E_L} \right) - \frac{1}{8} \left(\frac{E_{J1}}{E_L} \right)^2 + \dots \right] \\ E_{J2,\text{eff}} &= E_{J1} \left[-\gamma - \frac{1}{4} \left(\frac{E_{J1}}{E_L} \right) + \gamma \left(\frac{E_{J1}}{E_L} \right)^2 + \dots \right] \\ E_{J3,\text{eff}} &= E_{J1} \left[\gamma \left(\frac{E_{J1}}{E_L} \right) + \frac{1}{8} \left(\frac{E_{J1}}{E_L} \right)^2 + \dots \right]. \end{aligned} \quad (\text{S35})$$

Similar to the previous sections, truncating the summation at the second harmonic and retaining the leading order with respect to both γ and E_{J1}/E_L yields

$$U \approx -E_{J1} \cos(\varphi) + \underbrace{E_{J1} \left(\gamma + \frac{E_{J1}}{4E_L} \right)}_{E_{J2,\text{eff}}} \cos(2\varphi), \quad (\text{S36})$$

where the effective second harmonic appears as a sum of the two sources as discussed in the main text. For notational simplicity, the subscript “eff” will be omitted henceforth.

D. Effect of series inductance in classical picture

In this section, we briefly discuss the role of the series inductance through the classical plasma frequency of the SQUID. Assuming that the potential of a Josephson junction in series with an inductance is given by Eq. S19, the resulting SQUID potential can be written as

$$U_{\text{SQUID}}(\varphi; \phi_{\text{ext}}) = -E_{J1}^{(L)} \cos(\varphi) + \frac{E_{J1}^{(L)}}{4E_L} \cos(2\varphi) \quad (\text{S37})$$

$$-E_{J1}^{(R)} \cos(\varphi - \phi_{\text{ext}}) + \frac{E_{J1}^{(R)}}{4E_L} \cos(2(\varphi - \phi_{\text{ext}})). \quad (\text{S38})$$

The classical plasma frequency is obtained by evaluating the second derivative of the potential at its global minimum φ_{min} , which corresponds to approximating the potential by its quadratic expansion:

$$\omega_p = \sqrt{8E_C \left(\frac{d^2 U(\varphi)}{d\varphi^2} \Big|_{\varphi=\varphi_{\text{min}}} \right)}. \quad (\text{S39})$$

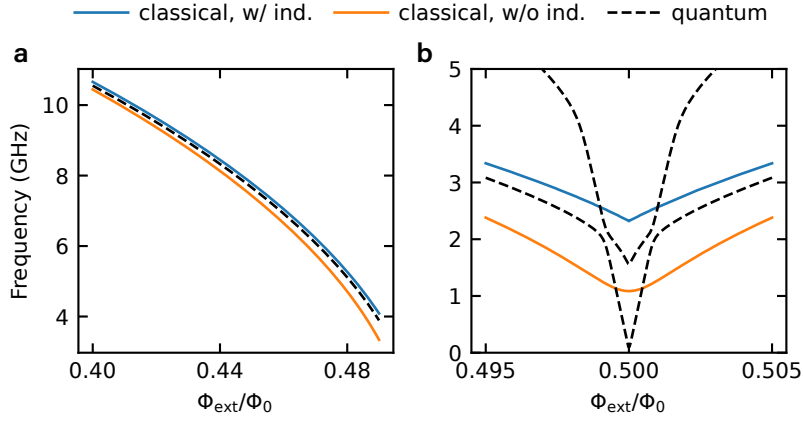


FIG. S6. Classical plasma frequency of the SQUID in the presence (blue) and absence (orange) of the series inductance. Transition frequency derived from a full-quantum mechanical approach (black dashed) is shown together.

Figure S6 compares the plasma frequencies of the SQUID calculated with and without the series inductance in each SQUID arm. The actual transition frequency obtained from a full quantum-mechanical approach is shown together. As seen in Fig. S6a, up to an offset caused by the higher-order potential terms beyond the quadratic approximation, similar to those that give rise to the standard $-E_C$ correction of the transmon frequency, the classical plasma frequency with the series inductance well predicts the overall trend of the actual transition away from half flux. Near half flux, the classical approach deviates as the quadratic approximation of the potential does not fully capture the double-well behavior. Nevertheless, this agreement implies that the harmonic correction to the current-phase relation due to the series inductance can already be anticipated within a classical picture.

S3. DEVICE TRANSITION SPECTRUM MEASUREMENT

A. Fitting algorithms

The device Hamiltonian under the second-harmonic approximation is

$$H = 4E_C \hat{n}^2 - E_{J1}^{(L)} \cos \hat{\varphi} - E_{J1}^{(R)} \cos (\hat{\varphi} - \phi_{\text{ext}}) + E_{J2}^{(L)} \cos (2\hat{\varphi}) + E_{J2}^{(R)} \cos (2(\hat{\varphi} - \phi_{\text{ext}})), \quad (\text{S40})$$

where the second harmonic $E_{J2}^{(i)}$ ($i = L, R$) is given by

$$E_{J2}^{(i)} = E_{J2,A}^{(i)} + E_{J2,L}^{(i)} = \gamma E_{J1}^{(i)} + \frac{(E_{J1}^{(i)})^2}{4E_L}. \quad (\text{S41})$$

Let $\alpha^{(i)}$ denote the ratio between the fundamental and the second harmonics:

$$\alpha^{(i)} = \frac{E_{J2}^{(i)}}{E_{J1}^{(i)}} = \gamma + \frac{E_{J1}^{(i)}}{4E_L}. \quad (\text{S42})$$

Then, we can re-express the Hamiltonian as

$$\hat{H} = 4E_C \hat{n}^2 - E_{J1}^{(L)} \left[\cos \hat{\varphi} - \alpha^{(L)} \cos (2\hat{\varphi}) \right] - E_{J1}^{(R)} \left[\cos (\hat{\varphi} - \phi_{\text{ext}}) - \alpha^{(R)} \cos (2\hat{\varphi} - 2\phi_{\text{ext}}) \right]. \quad (\text{S43})$$

Additionally, we introduce a new variable, dE_J , to represent the asymmetry between the two junctions of the left and right arms:

$$E_{J1}^{(R)} = E_{J1}^{(L)} (1 - dE_J). \quad (\text{S44})$$

$\alpha^{(L)}$ and $\alpha^{(R)}$ satisfy the following equation

$$\alpha^{(R)} = \gamma + \frac{E_{J1}^{(R)}}{4E_L} = \gamma + \frac{E_{J1}^{(L)} - E_{J1}^{(L)} dE_J}{4E_L} = \alpha^{(L)} - \frac{E_{J1}^{(L)}}{4E_L} dE_J. \quad (\text{S45})$$

To reduce the number of fit parameters, and given nominally identical junctions ($dE_J \ll 1$), we use the same ratio α for both arms:

$$\alpha^{(R)} \approx \alpha^{(L)} = \alpha, \quad (\text{S46})$$

which yields the fit Hamiltonian

$$H_{\text{fit}} = 4E_C \hat{n}^2 - E_{J1}^{(L)} \left[\cos \hat{\varphi} - \alpha \cos (2\hat{\varphi}) \right] - E_{J1}^{(L)} (1 - dE_J) \left[\cos (\hat{\varphi} - \phi_{\text{ext}}) - \alpha \cos (2\hat{\varphi} - 2\phi_{\text{ext}}) \right]. \quad (\text{S47})$$

We optimize the four circuit parameters, $\mathbf{x} = (E_C, E_{J1}^{(L)}, dE_J, \alpha)^T$, by minimizing the following cost function:

$$\mathbf{x}^* = \underset{E_C, E_{J1}^{(L)}, dE_J, \alpha}{\text{argmin}} \sum_i \sum_{\phi_{\text{ext}}} \left| \lambda_i (\phi_{\text{ext}}; E_C, E_{J1}^{(L)}, dE_J, \alpha) - f_i (\phi_{\text{ext}}) \right|^2, \quad (\text{S48})$$

where λ_i represents the i -th eigenenergy of the Hamiltonian in Eq. (S47), with the ground state ($i = 0$) energy set to zero, and f_i denotes the measured i -th transition frequency.

B. Details on spectrum measurement

As shown in Fig. S7 we measure two flux-bias ranges for the fit: one near $\Phi_{\text{ext}} = 0.5\Phi_0$, where the first transition frequency is approximately less than 3 GHz, and one farther away from $0.5\Phi_0$, where the first transition frequency falls within the 4 – 7 GHz range.

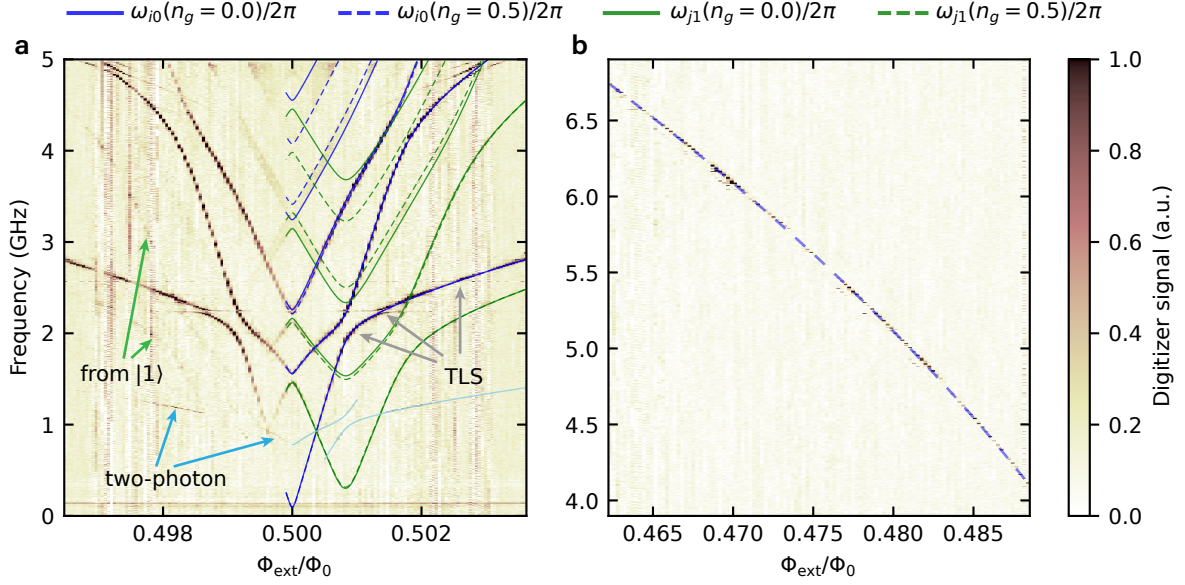


FIG. S7. Measured spectra of the 6 μm device for Chip A (a) near and (b) away from half flux.

In Fig. S7a, additional features apart from the main transitions from the ground state (blue) — such as the two-photon transition to the first plasmon state (sky blue), transitions from the first excited state (green), and the charge dispersions for both transitions from the ground state and the first excited state (solid and dashed lines)— are well captured by the second-harmonic model.

S4. ANALYSIS OF FULL QUBIT-RESONATOR SYSTEM

A. Effect of coupling to resonator

As shown in Section S3 A, we fit the measured spectra only using the circuit parameters of the capacitively shunted SQUID. However, coupling to the readout resonator may induce a frequency shift in the device. In this section, we discuss the effect of this coupling on the possible frequency shifts. Given the device Hamiltonian H_{fit} in Eq. (S47), the total system Hamiltonian including the resonator is given by

$$H_{\text{sys}} = H_{\text{fit}} + \hbar\omega_r \hat{a}^\dagger \hat{a} + \hbar g_c \hat{n} (\hat{a} - \hat{a}^\dagger) / i, \quad (\text{S49})$$

where ω_r is the bare resonator frequency, $\hat{a}$ is the annihilation operator of the resonator, and g_c is the coupling strength between the device and the resonator. As discussed in Fig. 3 (d)-(f) in the main text, g_c is separately calculated from the resonator single-tone spectroscopy.

In Fig. S8, we show the transition spectra for the Chip A devices with JJ lengths of $6\ \mu\text{m}$ (a-b) and $2\ \mu\text{m}$ (c-d) in the absence (orange solid) and presence (black dashed) of the resonator coupling. The former is obtained by diagonalizing Eq. (S47), and the latter from the system Hamiltonian in Eq. (S49). The energy eigenstates $|\tilde{l}\rangle$ of the system Hamiltonian for Fig. S8 were chosen based on their maximum overlap with the bare states: $\tilde{l} = \text{argmax}_{\tilde{k}} |\langle \tilde{k} | (|0\rangle_r \otimes |l\rangle_q) |$, where $|0\rangle_r$ is the ground state of the bare resonator, and $|l\rangle_q$ ($l \geq 0$) denotes the l th bare eigenstate of the device. We observe that the coupling strength is weak enough to cause negligible shift in the bare device spectra, indicating that the small coupling to the resonator does not change the fit parameters considerably.

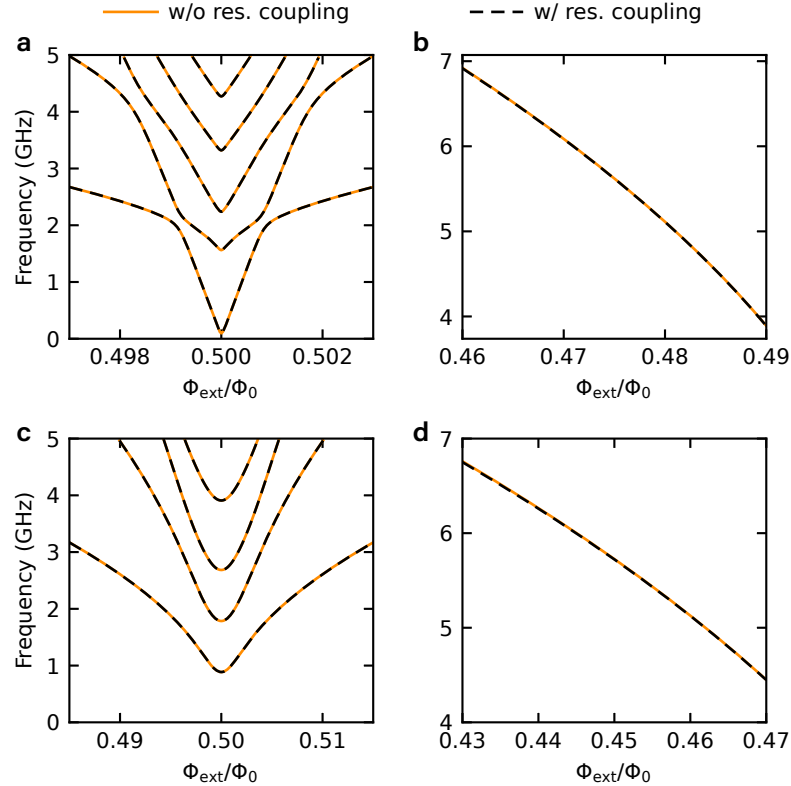


FIG. S8. Transition spectrum in the absence (orange solid) and presence (black dashed) of the interaction with the readout resonator. (a),(b) $6\ \mu\text{m}$ device (c),(d) $2\ \mu\text{m}$ device for Chip A. We find the frequency shifts of the device due to the interaction are negligible in our experiment.

Secondly, we discuss the applicability of the standard resonator frequency shift formula [Eq. (6) in the main text] to our circuit by comparing it with direct numerical diagonalization of the system Hamiltonian in Eq. (S49). We first

find two eigenstates $|\tilde{m}_0\rangle$ and $|\tilde{m}_1\rangle$ that maximize the overlap with the bare resonator states:

$$\begin{aligned}\tilde{m}_0 &= \operatorname{argmax}_{\tilde{k}} |\langle \tilde{k} | (|0\rangle_r \otimes |0\rangle_q) | \\ \tilde{m}_1 &= \operatorname{argmax}_{\tilde{k}} |\langle \tilde{k} | (|1\rangle_r \otimes |0\rangle_q) |.\end{aligned}$$

Next, we calculate the resonator frequency $\tilde{\omega}_r$ from the energy difference between the two states $\hbar\tilde{\omega}_r = E_{|\tilde{m}_1\rangle} - E_{|\tilde{m}_0\rangle}$. Fig. S9a shows that the resonator frequency shift obtained from standard second-order perturbation theory [Eq. (6) in the main text] agrees well with the result from direct diagonalization. As shown in Fig. S9b, although the perturbative result diverges at the exact bias point where the bare device transition frequency crosses the bare resonator frequency, it accurately predicts the resonator shift even very close to the crossing.

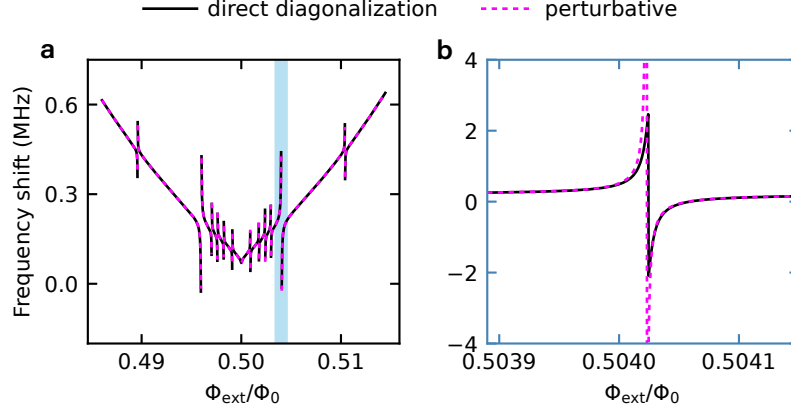


FIG. S9. (a) Comparison of the resonator frequency shift obtained from second-order perturbation theory (magenta dashed), as presented in the main text, and from direct diagonalization of Eq. (S49) (black solid) for the 6 μm device. The standard perturbative approach shows good agreement with the exact resonator frequency. (b) Resonator frequency shift near the avoided crossing (blue shaded region in (a)). Except for the bias point where the perturbative approach diverges, it accurately predicts the exact behavior of the resonator frequency.

B. Effect of AC stark shift

In this section, we discuss the AC Stark shift on the device transition spectra caused by the drive tone. In the spectroscopy measurement, the drive tone is applied through the feedline, which is inductively coupled to the readout resonator. Accordingly, the system Hamiltonian including the sinusoidal drive term can be modeled as

$$H' = H_{\text{sys}} + \Omega_{\text{rf}} (a + a^\dagger) B(t) \cos(\omega_d t) \quad (\text{S50})$$

where $B(t)$ is the time-dependent envelope of the drive tone, Ω_{rf} is the drive strength, and a is the resonator lowering operator. Note that the zero-point fluctuation of the resonator phase operator $\hat{\phi}_r = \phi_{\text{zpf}} (a + a^\dagger)$ is absorbed in Ω_{rf} .

To estimate Ω_{rf} , we first measure the Rabi oscillations of the $|\tilde{0}\rangle \rightarrow |\tilde{1}\rangle$ transition at a flux bias of $\Phi_{\text{ext}} = 0.5035 \Phi_0$, using a cosine envelope $B(t)$ defined as

$$B(t) = \frac{A}{2} [1 - \cos(2\pi t/t_g)],$$

where $t_g = 50$ ns is the gate time.

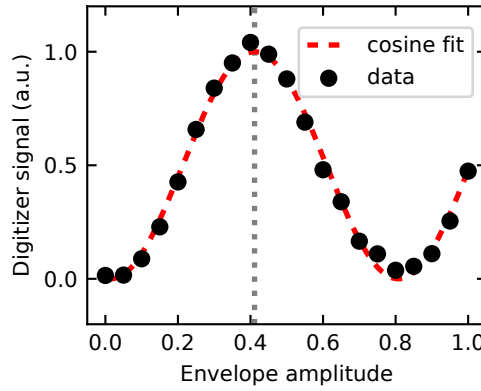


FIG. S10. Rabi oscillation as a function of envelope amplitude A .

Fig. S10 shows the measured Rabi oscillation as a function of the envelope amplitude A . The π -pulse amplitude is determined to be $A_\pi = 0.411$ (gray dashed line). Using this value, the drive strength for Rabi measurement $\Omega_{\text{rf}}^{\text{Rabi}}$ can be estimated from the following relation

$$\int_0^{t_g} \Omega_{\text{rf}}^{\text{Rabi}} |\langle \tilde{1} | a + a^\dagger | \tilde{0} \rangle| \frac{A_\pi}{2} [1 - \cos(2\pi t/t_g)] dt = \pi.$$

In the spectroscopy measurement, the applied RF power was 22 dB lower than in the Rabi measurement, satisfying $20 \log(\Omega_{\text{rf}}^{\text{spec}}/\Omega_{\text{rf}}^{\text{Rabi}}) = -22$. Therefore, in the spectroscopy measurement, the Hamiltonian in Eq. (S50) simplifies to

$$H' = H_{\text{sys}} + \Omega_{\text{rf}}^{\text{spec}} (a + a^\dagger) \cos \omega_d t, \quad (\text{S51})$$

where a constant envelope ($B(t) = 1$) is used, and $\Omega_{\text{rf}}^{\text{spec}}/2\pi = 860$ MHz is the estimated drive strength from the above calculation.

When the drive is on resonance with the transition, the AC Stark shift on the i -th energy ΔE_i^{AC} can be calculated using second-order perturbation theory applied to Eq. (S51) [5], yielding the following expression

$$\Delta E_i^{\text{AC}} = \frac{\hbar \Omega^2}{4} \sum_{j \neq i, |\omega_{ij}| \neq \omega_d} |\langle \tilde{j} | (a + a^\dagger) | \tilde{i} \rangle|^2 \left[\frac{1}{\tilde{\omega}_{ij} - \omega_d} + \frac{1}{\tilde{\omega}_{ij} + \omega_d} \right], \quad (\text{S52})$$

where $|\tilde{i}\rangle$ is the i -th energy eigenstate of H_{sys} , and $\tilde{\omega}_{ij} = \tilde{\omega}_i - \tilde{\omega}_j$ is the corresponding transition frequency. From Eq. (S52), the change in the transition frequency due to the AC Stark shift is estimated to be below 200 kHz for the sweep near half-flux [Fig. S7a] and below 12 MHz for the sweep farther from half-flux [Fig. S7b], which is negligible within the measured frequency window.

S5. SUPERCURRENT DIODE EFFECT WITH TRACE INDUCTANCE

In this section, we discuss supercurrent diode effect estimated in our device. In the presence of the second harmonic in the SQUID potential, the time-reversal symmetry can be broken when the device is biased at non-trivial flux $\Phi_{\text{ext}} \neq n\Phi_0/2$, $n \in \mathbb{Z}$ [6, 7]. In this case, the maximum and minimum supercurrents flowing across the SQUID become unbalanced, resulting in the diode effect. The effectiveness of the supercurrent diode is often quantified by the rectification efficiency η defined as the contrast between the maximum and minimum supercurrents [6]

$$\eta = \frac{|I_{\text{max}}| - |I_{\text{min}}|}{|I_{\text{max}}| + |I_{\text{min}}|} \quad (\text{S53})$$

Figure S11 shows the diode effect in the 6 μm device on Chip A. When biased close to half flux, the potential $U(\varphi)$ of the SQUID, calculated from Eq.(3) in the main text, becomes asymmetric due to the second harmonic [Fig. S11a]. The derivative of the potential with respect to the phase φ corresponds to the current through the SQUID. The difference in magnitudes of the maximum and minimum currents [Fig. S11b] features the presence of the diode effect in our device. As shown in Fig. S11c, our device is estimated to exhibit a rectification efficiency exceeding 10% near half flux.

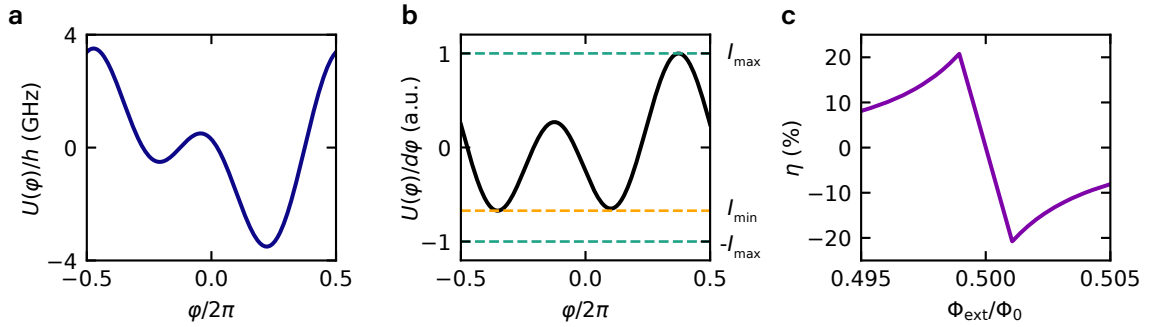


FIG. S11. (a) Potential energy of the 6 μm device on Chip A at $\Phi_{\text{ext}} \approx 0.499 \Phi_0$. (b) Supercurrent across the SQUID estimated from the derivative of the SQUID potential. The difference in I_{max} (green dashed) and I_{min} (orange dashed) indicate the diode effect. (c) Estimated rectification efficiency near half flux. The maximum efficiency is calculated to be over 20%.

Therefore, our results experimentally demonstrate that when engineering the diode effect using series and parallel combinations of JJs, for example, methods proposed in Refs. [7–9], the harmonic corrections arising from the embedding circuitry can be significant and must be carefully considered. We note that this consideration is not limited to the SIS tunnel junctions used in our work and is generally applicable to different types of JJs, for example, in an all-metallic junction as demonstrated in Ref. [6].

S6. INDUCTANCE SIMULATION

To estimate the inductance of the SQUID loop, we used the electromagnetic simulation package *InductEx*. As shown in Fig. S12a-S12b, we simulate the Dolan bridge region including the SQUID loop and two extended interconnects on the side.

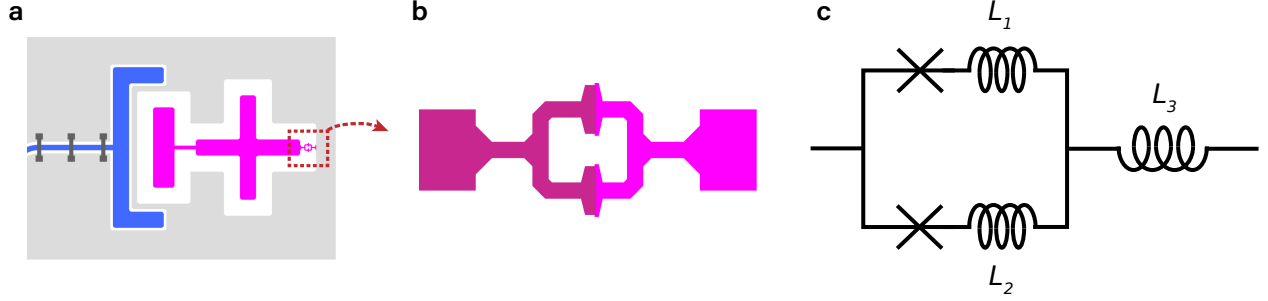


FIG. S12. (a) Full device geometry. (b) Simulated Dolan bridge region. The dark and bright colors represent the two shadow-evaporation layers. (c) Equivalent circuit model of the Dolan bridge.

The circuit model of the Dolan bridge is illustrated in Fig. S12c, where the loop inductance is modeled as two arms (L_1 , L_2), and the inductance of the side interconnects is in series with the loop (L_3). Note that the inductance on the left side of each junction is lumped into L_1 and L_2 since they are in series. Similarly, the bridge inductances on the two sides of the loop are lumped into L_3 . Using the actual device geometry [Fig. S12b] and the corresponding circuit model [Fig. S12c], the inductance values for each element of the circuit model were computed. The simulation results for five devices are summarized in Table S6. As discussed in Section S2 B, including the interconnect inductance did not significantly change the result of predicted qubit spectra, and is therefore omitted in the qubit energy calculation.

TABLE SIV. Simulated loop inductances (L_1 , L_2) and the interconnect inductance (L_3) for the five devices (indexed by nominal JJ lengths).

JJ length (μm)	2	3	4	5	6
L_1 (pH)	10.7	10.5	10.4	10.3	10.2
L_2 (pH)	10.7	10.5	10.4	10.3	10.2
L_3 (pH)	22.5	22.2	21.9	21.7	21.5

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