Supplementary material for energy decay and frequency shift of a superconducting qubit from non-equilibrium quasiparticles

M. Lenander[1], H. Wang[1,2], Radoslaw C. Bialczak[1], Erik Lucero[1], Matteo Mariantoni[1], M. Neeley[1], A. D. O'Connell[1], D. Sank[1], M. Weides[1], J. Wenner[1], T. Yamamoto[1,3], Y. Yin[1], J. Zhao[1], A. N. Cleland[1], and John M. Martinis[1]
[1] *Department of Physics, University of California, Santa Barbara, CA 93106, USA*
[2] *Department of Physics, Zhejiang University, Hangzhou 310027, China and*
[3] *Green Innovation Research Laboratories, NEC Corporation, Tsukuba, Ibaraki 305-8501, Japan*
(Dated: December 31, 2010)

Supplementary material is presented for the paper "Energy decay and frequency shift of a superconducting qubit from non-equilibrium quasiparticles". First, we discuss quasiparticle data for a superconducting coplanar resonator. We then document how the energy dependence of the occupation can be calculated numerically. We calculate analytically the total quasiparticle decay rate and time dependence with both a bulk model and, numerically, one including diffusion. We also compute the quasiparticle dependence of the gap, occupation probability, current-phase relationship, and how the frequency shift and dissipation are related. Finally, we calculate the Josephson current for non-equilibrium quasiparticles.

PACS numbers: 74.81.Fa, 03.65.Yz, 74.25.Nf, 74.50.+r

We are interested in how non-equilibrium quasiparticles affect the properties of a Josephson junction in qubit devices. Since we are concerned with very low temperature operation, we consider phonon temperatures sufficiently low that no quasiparticles exist due to thermal generation. The non-equilibrium quasiparticles are generated with an unknown mechanism, and then relax their energy via the emission of phonons. The quasiparticles typically have energy E close to the gap, from which they eventually decay via recombination. From electron-phonon physics, we know that the qusiaparticles relax to energies very near the gap, as calculated in Ref. [4].

The factor of 2 in the definition of n_{qp} comes from integration only over positive energy, whereas excitations arise from both electron states above and below the Fermi energy. Note this integral already contains the two possible spin states in the definition of $D(E_f)$.

QUASIPARTICLES IN COPLANAR RESONATORS

The relationship between quasiparticle damping and frequency shift may also be tested in superconducting resonators. Here microwave transmission is measured to extract the resonance frequency f and the quality factor Q, with the quasiparticle density changed by simply increasing the temperature [1]. As shown in Fig. 1, we find that with increasing quasiparticle density, dissipation increases and the resonance frequency decreases, in a similar manner as for junctions.

To compare with theory, we use solutions to the Mattis-Bardeen conductivity that is valid for the regime $kT \sim \hbar\omega \ll \Delta$ [2, 3]. These results can be expressed in terms of an admittance function discussed in the main article, but with a modification of the the 1+i term

$$\frac{Y_j(\omega)}{1/R_n} = (d_R + i d_I)\sqrt{2}\left(\frac{\Delta}{\hbar\omega}\right)^{3/2}\frac{n_{\mathrm{qp}}}{n_{\mathrm{cp}}} - i\pi\frac{\Delta}{\hbar\omega}\ , \quad (1)$$

$$d_R = 2\sqrt{2x/\pi}\,\sinh(x)K_0(x)\ , \quad (2)$$

$$d_I = \sqrt{2\pi x}\,\exp(-x)I_0(x)\ , \quad (3)$$

where $x = \hbar\omega/2kT$. The theoretical prediction, indicated by the gray line in Fig. 1, is in reasonable agreement with the data at high temperatures. The experimental slope at temperatures above 200 mK is 0.77 times that given by theory. The deviation is largest below about 120 mK, and believed to arise from two-level states that are not included in this model.

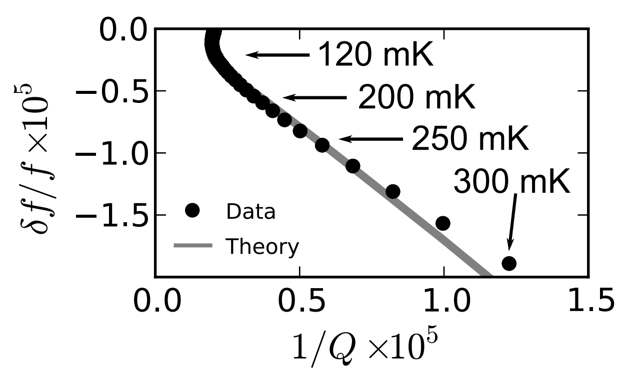

FIG. 1: Parametric plot of fractional frequency shift $\delta f/f$ versus dissipation $1/Q$, for various quasiparticle occupations n_{qp} changed by varying the the sample temperature. The device is a aluminum coplanar resonator fabricated on sapphire. The slope of the data is in good agreement with predictions (gray line) at temperatures above 250 mK. Deviations at low temperature are believed to come from the two-level states.

NUMERICAL SOLUTION OF QUASIPARTICLE RECOMBINATION

For numerical computations of non-equilibrium quasiparticle density from relaxation and recombination [4], the integrals over energy have to be put into discrete form. For a binning size given by $d\epsilon$ in energy, we define the number of excitations in bin i as

$$n_i \equiv d\epsilon\rho(\epsilon_i)f(\epsilon_i)\ . \tag{4}$$

Using this definition, the total quasiparticle density is

$$\frac{n_{\rm qp}}{n_{\rm cp}} = \frac{2}{\Delta}\sum_i n_i \tag{5}$$

The scattering and recombination rates of Eqs. (6) and (7) of Ref. [4] can then be expressed in a discrete form

$$\Gamma^s_{i\to j} = \sum_j \Big[\frac{(\epsilon_i-\epsilon_j)^2}{\tau_0(kT_c)^3}(1-\frac{\Delta^2}{\epsilon_i\epsilon_j})N_p(\epsilon_i-\epsilon_j)d\epsilon\rho(\epsilon_j)\Big] \tag{6}$$

$$\equiv \sum_j G^s_{ij}\ , \tag{7}$$

$$\Gamma^r_{i,j} = \sum_j \Big[\frac{(\epsilon_i+\epsilon_j)^2}{\tau_0(kT_c)^3}(1+\frac{\Delta^2}{\epsilon_i\epsilon_j})N_p(\epsilon_i+\epsilon_j)\Big]d\epsilon\rho(\epsilon_j)f(\epsilon_j) \tag{8}$$

$$\equiv \sum_j G^r_{ij}\ n_j\ , \tag{9}$$

where the phonon occupation factor is $N_p(E) = 1/|\exp(-E/kT_p)-1|$ and the bracketed terms are the G factors. We have also assumed small occupation, so that in Eq. (6) we use $1-f\to 1$.

The coupled differential equations for the change in the excitation number are

$$\frac{d}{dt}n_i = G^s_{ji}n_j - \sum_j G^s_{ij}n_i - \sum_j(1+\delta_{ij})G^r_{ij}n_jn_i\ , \tag{10}$$

where δ_{ij} is the Kronecker delta and accounts for the annihilation of 2 quasiparticles when in the same bin ($i=j$). With the physics expressed in matrix form, a solution can be readily solved numerically.

QUASIPARTICLE DECAY

The physics of quasiparticle relaxation and recombination was discussed in Ref. [4]. Although the article described solutions for the non-equilibrium occupation $f(E)$ using numerical methods, quasiparticle decay physics can be understood in the case of low density where they are mostly occupied at the gap. The electron-electron recombination rate of a single quasiparticle, starting from Eq. (7) of Ref. [4], can be well approximated using

$$\Gamma^r \simeq \frac{1}{\tau_0}\int_\Delta^\infty d\epsilon'\frac{(\epsilon+\epsilon')^2}{(kT_c)^3}(1+\frac{\Delta^2}{\epsilon\epsilon'})\rho(\epsilon')f(\epsilon') \tag{11}$$

$$\simeq \frac{1}{\tau_0}\frac{(2\Delta)^2}{(kT_c)^3}(1+\frac{\Delta^2}{\Delta^2})\int_\Delta^\infty d\epsilon'\rho(\epsilon')f(\epsilon') \tag{12}$$

$$= \frac{4}{\tau_0}(1.76)^3\frac{n_{\rm qp}}{D(E_F)\Delta} \tag{13}$$

$$= \frac{21.8}{\tau_0}\frac{n_{\rm qp}}{n_{\rm cp}}\ , \tag{14}$$

where we have used the BCS result $\Delta/kT_c = 1.76$. Here, $D(E_f)/2$ is the single-spin density of states, and we define the Cooper pair density $n_{\rm cp} \equiv D(E_F)\Delta$.

The time dependence of the quasiparticle density can be understood via the rate equation

$$\frac{d}{dt}n_{\rm qp} = -2\Gamma^r n_{\rm qp} + r_{\rm qp} \tag{15}$$

$$\frac{d}{dt}\frac{n_{\rm qp}}{n_{\rm cp}} = -\frac{43.6}{\tau_0}\Big(\frac{n_{\rm qp}}{n_{\rm cp}}\Big)^2 + \frac{r_{\rm qp}}{n_{\rm cp}}\ , \tag{16}$$

where a recombination event removes 2 quasiparticles, $r_{\rm qp}$ is the single particle quasiparticle injection rate. The second equation is for the normalized quasiparticle density, and has a recombination rate that is proportional to $n^2_{\rm qp}$ because of the two-body electron-electron interaction.

The equilibrium quasiparticle density is given by setting $dn_{\rm qp}/dt = 0$, yielding density and recombination rates

$$\frac{(n_{\rm qp})^{\rm eq}}{n_{\rm cp}} = \Big[\frac{\tau_0}{43.6}\frac{r_{\rm qp}}{n_{\rm cp}}\Big]^{1/2} = \frac{\tau_0}{43.6}(\Gamma^r)^{\rm eq}\ , \tag{17}$$

$$(\Gamma^r)^{\rm eq} = \Big[\frac{43.6}{\tau_0}\frac{r_{\rm qp}}{n_{\rm cp}}\Big]^{1/2} = \frac{43.6}{\tau_0}\frac{(n_{\rm qp})^{\rm eq}}{n_{\rm cp}}\ . \tag{18}$$

The first equation is close to what was found numerically in Ref. [4]. The second is given by the geometric mean of the normalized injection and the characteristic electron-electron interaction rates.

We compared the results of this simple calculation with numerical solutions for a range of injection rates and found excellent agreement for $n_{\rm qp}/n_{\rm cp} \lesssim 0.001$. Even at large density $n_{\rm qp}/n_{\rm cp} = 0.1$, Eq. (17) is a reasonable approximation as its prediction is only 40 % larger than that obtained via numerics.

For no injection of quasiparticles $r_{\rm qp} = 0$, the differential equation can be integrated to give

$$\frac{n_{\rm qp}}{n_{\rm cp}} = \frac{\tau_0/43.6}{t-t_0} \tag{19}$$

where t is the time and t_0 is an integration constant, which is approximately the time at which the quasiparticles start to cool. The solution to the differential equation for a finite injection rate is

$$n_{\rm qp} = (n_{\rm qp})^{\rm eq}\coth[\,(\Gamma^r)^{\rm eq}(t-t_0)\,]\ , \tag{20}$$

where the coth term is replaced by tanh if the quasiparticle density increases with time. At short times the term $\coth\Gamma^r t = 1/\Gamma^r t$, which then gives Eq. (19) and a time dependence that scales *only* with the electron-phonon coupling time τ_0. There is a relatively sharp crossover to the long time behavior where the quasiparticle density $n_{\rm qp}^{\rm eq}$ is constant with time. The crossover time is given by $1/(\Gamma^r)^{\rm eq}$.

The inverse of the crossover time thus gives the equilibrium recombination rate $(\Gamma^r)^{\rm eq}$, which is related to the density using Eq. (17) and the parameter τ_0. Comparing this density with that found from the qubit decay rate allows one to determine whether quasiparticles are the limiting decay mechanism for the qubit.

QUASIPARTICLE DECAY WITH DIFFUSION

The analysis in the last section assumes a bulk (uniform) model where there is no diffusion of quasiparticles. Here, we describe a numerical solution for quasiparticle decay including relaxation, recombination, and diffusion using the simple geometry of a thin superconducting disk of radius 5 mm. We use constant quasiparticle injection throughout the disk and a large injection pulse into the center of the disk at time $t = -200\,\mu$s to $t = 0$. Because diffusion depends on the quasiparticle energy, the calculation keeps track of the occupation probability for both the radius and energy variables.

In Fig. 2 we plot quasiparticle density versus settling time in a manner similar to that in the main paper, but for 4 radii. We find differing behavior depending on the ratio of the radius with the diffusion length $\sim 1\,$mm, as computed for e-e diffusion in Fig. 3 of Ref. [4]. For small radii, we see a dependence on time that matches closely with the bulk theory, as described in the main paper. For a radius much larger than the diffusion length, the quasiparticle density does not change. For the radius close to the diffusion length, we observed behavior between the two limits - a reduced peak density but a relaxation to the steady state value that has a similar time scale than for a small radius.

We note that the actual qubit device has interruptions in the ground plane due to the device geometry, so that this computation will not exactly match the experimental data. However, the model mimics the time dependence of the data fairly well above $10\,\mu$s, so it is reasonable to compare to the simple bulk analysis for the behavior at long times $\gtrsim 250\,\mu$s.

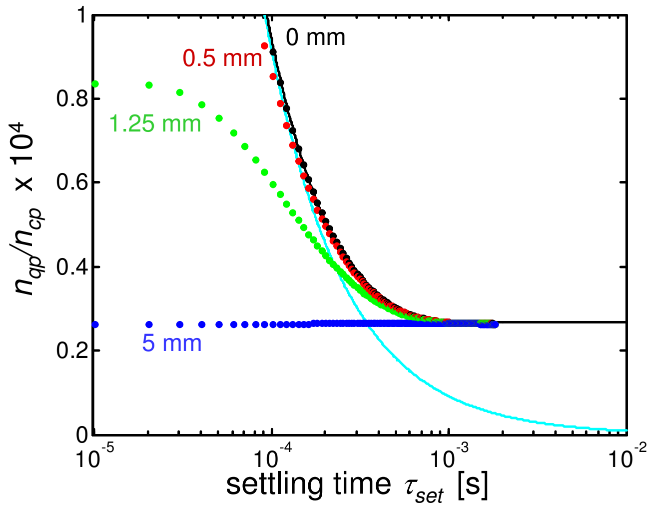

FIG. 2: Plot of quasiparticle density versus settling time for a superconducting disk of radius 5 mm with quasiparticle injection at the center. Points are for the simulation at four radii, r = 0 (black), 0.5 mm (red), 1.25 mm (green), and 5 mm (blue). Large changes are observed for small radii, which show a time dependence close to that predicted by the full theory of Eq. (9) of main article (black line) and for the zero background of Eq. (8) (cyan line). At large radii much greater than the characteristic diffusion length of $\sim 1\,$mm, no change in quasiparticle density is seen. The simulation for radius 1.25 mm is in reasonable agreement with experimental data.

DEPENDENCE OF GAP ON QUASIPARTICLES

The change in the superconducting gap Δ with quasiparticles can be calculated starting from the BCS gap equation, but assuming a small non-equilibrium population $f(E)$

$$\Delta = D(E_f)V\int_\Delta^{\theta_D} dE\,\rho\frac{\Delta}{E}(1-2f)\ , \tag{21}$$

$$1 = D(E_f)V\Big(\int_\Delta^{\theta_D}\frac{dE}{\sqrt{E^2-\Delta^2}} - \int_\Delta^{\theta_D} dE\,\rho\frac{1}{E}2f\Big) \tag{22}$$

$$\simeq D(E_f)V\Big(\log\frac{2\theta_D}{\Delta} - \frac{n_{\rm qp}}{D(E_f)\Delta}\Big) \tag{23}$$

where V is the attraction potential and θ_D is the Debye energy. Solving for the gap, one finds

$$\Delta = 2\theta_D\exp\Big(-\frac{1}{D(E_F)V} - \frac{n_{\rm qp}}{D(E_F)\Delta}\Big) \tag{24}$$

$$= \Delta_0\exp\Big(-\frac{n_{\rm qp}}{D(E_F)\Delta}\Big) \tag{25}$$

$$\simeq \Delta_0\Big(1-\frac{n_{\rm qp}}{n_{\rm cp}}\Big)\ , \tag{26}$$

where Δ_0 is the normal expression for the BCS gap with no quasiparticles.

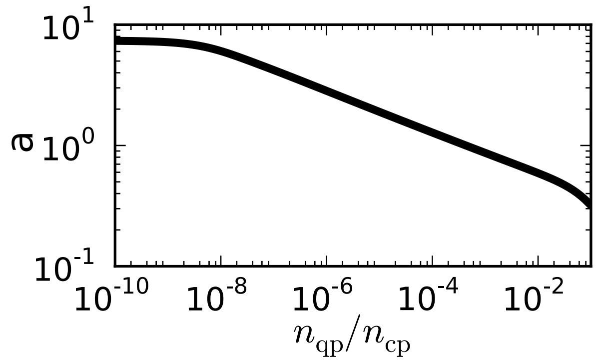

FIG. 3: Plot of $a = f(\Delta)/(n_{\rm qp}/n_{\rm cp})$ versus $n_{\rm qp}/n_{\rm cp}$, found from numerical simulation. Based on the values of $n_{\rm qp}/n_{\rm cp}$ in Fig. 2b in the paper, we find $a \simeq 1.2$. For a wide range of injection rates, a can be well approximated by the power law $a \simeq 0.12\,(n_{\rm qp}/n_{\rm cp})^{-0.173}$.

NUMERICAL DETERMINATION OF OCCUPATION PARAMETER

To determine the effect of non-equilibrium quasiparticles, both the quasiparticle density $n_{\rm qp}$ and the occupation probability at the gap $f(\Delta)$ must be calculated. We plot in Fig. 3 the quantity $a = f(\Delta)/(n_{\rm qp}/n_{\rm cp})$ versus $n_{\rm qp}/n_{\rm cp}$ obtained from numerical computations for wide range of injection rates. We find the results are well approximated by a line on the log-log plot, implying that the dependence can be well approximated by the power-law formula $a \simeq 0.12\,(n_{\rm qp}/n_{\rm cp})^{-0.173}$.

CURRENT-PHASE RELATIONSHIP WITH QUASIPARTICLES

The current-phase relationship from Josephson tunneling is given by

$$I(\phi) = I_0 \sin\phi \Big[1 - \frac{n_{\rm qp}}{n_{\rm cp}}\Big]\big[1 - 2f(\Delta)\big]\ , \qquad (27)$$

where $I_0 = \pi\Delta_0/2eR_n$ and the dependence of Δ on quasiparticles is now explicitly shown. To first order, the fractional change in critical current is

$$\frac{\delta I_0}{I_0} = -(1+2a)\frac{n_{\rm qp}}{n_{\rm cp}}\ . \qquad (28)$$

An interesting question is whether the quasiparticle tunneling terms should also be included in the current-phase relation. For the junction current to only be a function of phase, it must arise for a purely inductive component of the junction admittance, which corresponds to terms with a frequency dependence that scales as $1/i\omega$. Tunneling of free quasiparticles should not be included since it has an additional frequency dependence $(n_{\rm qp}/n_{\rm cp})\sqrt{\Delta/\hbar\omega}$. The Andreev bound states of the quasiparticles have an inductance, so the AC Josephson relation can then be used to find the current

$$I_{ABS}(\phi) = -\frac{\Phi_0}{2\pi}\int_0^{\phi} {\rm Im}\{\omega Y_{ABS}(\phi)\}\ d\phi \qquad (29)$$

$$= I_0[\phi + \sin\phi] f(\Delta)\ . \qquad (30)$$

Note that this term increases the critical current, and if one replaces $\phi \to \sin\phi$ it cancels out the decrease coming from Josephson tunneling. Since many experiments have shown the temperature dependence of the current-phase relation is given by only Josephson tunneling, we do not use this Andreev bound state term in our calculations. In addition, our data is not consistent with including this term since it has the effect of reducing $2a$ in Eq. (28) to a value below a.

RELATING DISSIPATION AND THE FREQUENCY CHANGE

Since both dissipation and the fractional critical-current change are proportional to $n_{\rm qp}$, the magnitude of these effects are related. The fractional change in the qubit resonance frequency E_{10}/h can be calculated knowing that its dominant scaling is $E_{10}/h \propto (I_0 - I)^{1/4}$, where I is the qubit bias current, giving

$$\frac{\delta(E_{10}/h)}{E_{10}/h} = \frac{1}{4}\frac{\delta(I_0 - I)}{I_0 - I} \qquad (31)$$

$$= \frac{1}{4}\frac{I_0}{I_0 - I}\frac{\delta I_0}{I_0} \qquad (32)$$

$$= -\frac{1+2a}{4}\frac{I_0}{I_0 - I}\ \frac{n_{\rm qp}}{n_{\rm cp}}\ . \qquad (33)$$

Quasiparticle dissipation can be likewise written in terms of the quasiparticle density, provided we first re-express the capacitance C into qubit parameters. Using the Josephson inductance $L_{J0} = \Phi_0/2\pi I_0$, the qubit resonance frequency is given by

$$\frac{E_{10}}{h} \simeq \frac{1}{2\pi}\frac{1}{\sqrt{L_{J0}C}}\ [2(I_0 - I)/I_0]^{1/4}\ . \qquad (34)$$

We thus calculate the decay rate of the qubit

$$\Gamma_1 \simeq \frac{1+\cos\phi}{\sqrt{2}}\frac{2eI_0}{\pi\Delta C}\Big(\frac{\Delta}{E_{10}}\Big)^{3/2}\frac{n_{\rm qp}}{n_{\rm cp}} \qquad (35)$$

$$= \frac{1+\cos\phi}{\sqrt{2}}\frac{h}{2\pi^2\Delta}\frac{1}{L_{J0}C}\Big(\frac{\Delta}{E_{10}}\Big)^{3/2}\frac{n_{\rm qp}}{n_{\rm cp}} \qquad (36)$$

$$\simeq \frac{1+\cos\phi}{\sqrt{2}}\frac{h}{2\pi^2\Delta}\frac{(2\pi E_{10}/h)^2}{\sqrt{2(I_0 - I)/I_0}}\Big(\frac{\Delta}{E_{10}}\Big)^{3/2}\frac{n_{\rm qp}}{n_{\rm cp}} \qquad (37)$$

$$= (1+\cos\phi)\frac{E_{10}}{h}\Big(\frac{I_0}{I_0 - I}\Big)^{1/2}\Big(\frac{\Delta}{E_{10}}\Big)^{1/2}\frac{n_{\rm qp}}{n_{\rm cp}}\ . \qquad (38)$$

By taking the ratio of Eqs. (33) and (38), the quasiparticle densities cancel out, and we can relate the dissipation to the frequency shift

$$\frac{\delta(E_{10}/h)}{\Gamma_1} = -\frac{1}{4}\frac{1+2a}{1+\cos\phi}\Big(\frac{I_0}{I_0-I}\Big)^{1/2}\Big(\frac{E_{10}}{\Delta}\Big)^{1/2} \ . \quad (39)$$

JOSEPHSON EFFECT FOR ARBITRARY QUASIPARTICLE OCCUPATION

Here we calculate the effect of a non-equilibrium population of quasiparticle states on Josephson tunneling, as appropriate for qubit devices. The current proportional to $\cos\delta$ is also evaluated for the case of gaps that are not equal. The results are readily obtained using standard second-order perturbation theory and simple integration of intermediate formulas.

The work expands on Ref. [5], which calculated the Josephson effect at zero temperature. In the paper, the section on Josephson tunneling is the starting point of this calculation.

The Josephson effect is derived by calculating the second-order change in energy to a superconducting state from a tunnel junction. The tunneling Hamiltonian in second-order perturbation theory is given by

$$H_T^{(2)} = \sum_i H_T \frac{1}{\epsilon_i} H_T \ , \quad (40)$$

where ϵ_i is the energy of the intermediate state i. Because the terms in H_T have both $\gamma^\dagger$ and γ operators, the second-order Hamiltonian has terms that transfers charge across the junction but does not change the superconducting state, thus giving a change in the energy of the state. This differs from first-order tunneling theory, which produces current only through the real creation of quasiparticles.

Because H_T has terms that transfer charge in both directions, $H_T H_T$ will produce terms which transfer two electrons to the right, two to the left, and with no net transfer. With no transfer, a calculation of the second-order energy gives a constant value, which has no physical effect. We first calculate terms for the transfer of two electrons to the right from $(\overrightarrow{H}_{T+}+\overrightarrow{H}_{T-})(\overrightarrow{H}_{T+}+\overrightarrow{H}_{T-})$. Nonzero expectation values are obtained for only two out of the four terms, as given by

$$\overrightarrow{H_T^{(2)}} = \sum_i \frac{\overrightarrow{H}_{T+}\overrightarrow{H}_{T-}+\overrightarrow{H}_{T-}\overrightarrow{H}_{T+}}{\epsilon_i} \quad (41)$$

$$= \sum_i |t|^2 \frac{(c_L c_R^\dagger)(c_{-L}c_{-R}^\dagger)+(c_{-L}c_{-R}^\dagger)(c_L c_R^\dagger)}{\epsilon_i} \quad (42)$$

$$= \sum_i |t|^2 \frac{(c_L c_{-L})(c_R^\dagger c_{-R}^\dagger)+(c_{-L}c_L)(c_{-R}^\dagger c_R^\dagger)}{\epsilon_i} \quad (43)$$

The pairs of electron creation and annihilation operators can be computed, giving

$$c_k c_{-k} = (u\gamma_0 + ve^{i\phi}\gamma_1^\dagger)(u\gamma_1 - ve^{i\phi}\gamma_0^\dagger) \quad (44)$$

$$\to uve^{i\phi}(-\gamma_0\gamma_0^\dagger + \gamma_1^\dagger\gamma_1) \ , \quad (45)$$

$$c_{-k}c_k \to uve^{i\phi}(-\gamma_0^\dagger\gamma_0 + \gamma_1\gamma_1^\dagger) \ , \quad (46)$$

$$c_k^\dagger c_{-k}^\dagger = (u\gamma_0^\dagger + ve^{-i\phi}\gamma_1)(u\gamma_1^\dagger - ve^{-i\phi}\gamma_0) \quad (47)$$

$$\to uve^{-i\phi}(-\gamma_0^\dagger\gamma_0 + \gamma_1\gamma_1^\dagger) \ , \quad (48)$$

$$c_{-k}^\dagger c_k^\dagger \to uve^{-i\phi}(-\gamma_0\gamma_0^\dagger + \gamma_1^\dagger\gamma_1) \ , \quad (49)$$

where we have only included pairs of quasiparticle operators γ that leaves the superconducting state unchanged, as needed for a calculation of the energy change from tunneling. Inserting these operators into Eq. (43) and defining the phase difference $\delta = \phi_L - \phi_R$, we find

$$\overrightarrow{H_T^{(2)}} = \sum_i |t|^2 e^{i\delta}(u_L v_L)(u_R v_R)\frac{(-\gamma_{L0}\gamma_{L0}^\dagger + \gamma_{L1}^\dagger\gamma_{L1})(-\gamma_{R0}^\dagger\gamma_{R0} + \gamma_{R1}\gamma_{R1}^\dagger) + (-\gamma_{L0}^\dagger\gamma_{L0} + \gamma_{L1}\gamma_{L1}^\dagger)(-\gamma_{R0}\gamma_{R0}^\dagger + \gamma_{R1}^\dagger\gamma_{R1})}{\epsilon_i} \quad (50)$$

$$= \sum_i |t|^2 e^{i\delta}(u_L v_L)(u_R v_R)\Big[\frac{-\gamma_{L0}\gamma_{L0}^\dagger\gamma_{R1}\gamma_{R1}^\dagger - \gamma_{L1}\gamma_{L1}^\dagger\gamma_{R0}\gamma_{R0}^\dagger}{E_L+E_R} + \frac{-\gamma_{L1}^\dagger\gamma_{L1}\gamma_{R0}^\dagger\gamma_{R0} - \gamma_{L0}^\dagger\gamma_{L0}\gamma_{R1}^\dagger\gamma_{R1}}{-E_L-E_R}$$
$$+ \frac{\gamma_{L0}\gamma_{L0}^\dagger\gamma_{R0}^\dagger\gamma_{R0} + \gamma_{L1}\gamma_{L1}^\dagger\gamma_{R1}^\dagger\gamma_{R1}}{E_L-E_R} + \frac{\gamma_{L1}^\dagger\gamma_{L1}\gamma_{R1}\gamma_{R1}^\dagger + \gamma_{L0}^\dagger\gamma_{L0}\gamma_{R0}\gamma_{R0}^\dagger}{-E_L+E_R}\Big] \ , \quad (51)$$

where we have computed the intermediate energy ϵ_i using a positive (negative) energy E for the creation (annihilation) of a quasiparticle. The quantity $u\,v = \Delta/2E$ describes the amplitude for the virtual quasiparticle to be both electron- and hole-like, which allows a net transfer of charge by two electrons.

We now change the sum to an integral over electron states according to

$$\sum_i \to N_{0L}\int_{-\infty}^{\infty} d\xi_L \quad N_{0R}\int_{-\infty}^{\infty} d\xi_R \quad = 2N_{0L}\int_{\Delta_L}^{\infty}\rho_L\ dE_L \quad 2N_{0R}\int_{\Delta_R}^{\infty}\rho_R\ dE_R \quad , \tag{52}$$

where N_0 is the normal density of states, and $\rho = E/\sqrt{E^2-\Delta^2}$ is the (normalized) superconducting density of states.

By describing the superconducting state with an occupation probability of quasiparticles $f = f(E)$, the quasiparticle operators for the creation then destruction of a quasiparticle is weighted by $1-f$, while the process of destruction then creation is weighted by f. The tunneling Hamiltonian is then given by

$$\overrightarrow{H_T^{(2)}} = |t|^2 e^{i\delta} N_{0L}N_{0R}\ 2\int_{\Delta_L}^{\infty}\rho_L dE_L\ 2\int_{\Delta_R}^{\infty}\rho_R dE_R\ \frac{\Delta_L}{2E_L}\ \frac{\Delta_R}{2E_R}\ 2G \quad , \tag{53}$$

$$G = -\frac{(1-f_L)(1-f_R)}{E_L+E_R} - \frac{f_L f_R}{-E_L-E_R} + \frac{(1-f_L)f_R}{E_L-E_R+i\epsilon} + \frac{f_L(1-f_R)}{-E_L+E_R+i\epsilon} \tag{54}$$

$$= -\frac{1-f_L-f_R+f_Lf_R}{E_L+E_R} + \frac{f_Lf_R}{E_L+E_R} + \frac{f_R-f_Lf_R}{E_L-E_R+i\epsilon} + \frac{f_L-f_Lf_R}{-E_L+E_R+i\epsilon} \tag{55}$$

$$= -\frac{1-f_L-f_R}{E_L+E_R} + \mathrm{P}\frac{f_R-f_L}{E_L-E_R} + i\pi(f_L+f_R-2f_Lf_R)\delta(E_L-E_R) \tag{56}$$

$$= -\frac{1}{E_L+E_R} + \mathrm{P}\frac{2f_RE_L-2f_LE_R}{E_L^2-E_R^2} + i\pi(f_L+f_R-2f_Lf_R)\delta(E_L-E_R) \quad , \tag{57}$$

where G comes from quasiparticle operators (bracket terms in Eq. (51)) after removing a factor of 2 because of the pair of states 0 and 1. We take $\epsilon \to 0+$, and the integration over the zero of energy in the denominator is performed using $1/(x+i\epsilon) = \mathrm{P}(1/x) + i\pi\delta(x)$, where P is the principle part and $\delta(x)$ is the Dirac δ-function.

The total second-order Hamiltonian for the tunneling of 2 electrons in both directions is

$$H_T^{(2)} = \overrightarrow{H_T^{(2)}} + \overleftarrow{H_T^{(2)}} \tag{58}$$

$$= \overrightarrow{H_T^{(2)}} + \text{h.c.} \tag{59}$$

$$= 2\mathrm{Re}\{\overrightarrow{H_T^{(2)}}\} \quad , \tag{60}$$

where h.c. is the Hermitian conjugate.

This result can be expressed in more physical terms by noting that the junction resistance can be written as

$$\frac{1}{R_n} = \frac{4\pi e^2}{\hbar}|t|^2 N_{0R}N_{0L} \quad . \tag{61}$$

In addition, The Josephson tunneling current is given by

$$I_j = \frac{2e}{\hbar}\frac{\partial\langle H_T^{(2)}\rangle}{\partial\delta} \quad . \tag{62}$$

Combining all of these equations, the total Josephson current is given by the integrals

$$I_j = \frac{2}{\pi e R_n}\Big\{\sin\delta\ \mathrm{P}\int_{\Delta_L}^{\infty}\rho_L dE_L\int_{\Delta_R}^{\infty}\rho_L dE_R\ \frac{\Delta_L}{E_L}\ \frac{\Delta_R}{E_R}\Big[\frac{1}{E_L+E_R} - 2\frac{f_RE_L-f_LE_R}{E_L^2-E_R^2}\Big]$$
$$-\pi\cos\delta\int_{\max(\Delta_L,\Delta_R)}^{\infty}\rho_L\rho_R dE\ \frac{\Delta_L\Delta_R}{E^2}(f_L+f_R-2f_Lf_R)\Big\} \quad , \tag{63}$$

We note that the $\sin\delta$ term in Eq. (63) corresponds to Eq. (22) of the Ambegaokar-Baratoff calculation [6].

We evaluate these integrals by first considering, without loss of generality, that $\Delta_L < \Delta_R$. Using $\rho\Delta/E = \Delta/\sqrt{E^2-\Delta^2}$ and $y = E_R/\Delta_L > 1$, we compute that the temperature independent term $1/(E_L+E_R)$ gives for integration over E_L

$$I_{1L} = \int_{\Delta_L}^{\infty} dE_L\frac{\Delta_L}{\sqrt{E_L^2-\Delta_L^2}}\frac{1}{E_L+E_R} \tag{64}$$

$$= \int_1^{\infty} dx\frac{1}{\sqrt{x^2-1}}\ \frac{1}{x+y} \tag{65}$$

$$= \frac{\operatorname{arccosh} y}{\sqrt{y^2-1}} \quad . \tag{66}$$

The remaining integration over E_R gives [7]

$$I_{1LR} = \int_{\Delta_R}^{\infty} dE_R \frac{\Delta_R}{\sqrt{E_R^2-\Delta_R^2}} \; \frac{\Delta_L \text{arccosh}(E_R/\Delta_L)}{\sqrt{E_R^2-\Delta_L^2}} \tag{67}$$

$$= \pi \frac{\Delta_L\Delta_R}{\Delta_L+\Delta_R} \text{EllipticK}\Big|\frac{\Delta_L-\Delta_R}{\Delta_L+\Delta_R}\Big| \tag{68}$$

$$= \frac{\pi^2}{4}\Delta \qquad \text{(for } \Delta_L=\Delta_R=\Delta), \tag{69}$$

where the last equation uses EllipticK(0) $= \pi/2$. From numerical integration, we have found that Eq. (68) is only approximate for $\Delta_L \neq \Delta_R$.

For the next term in Eq. (63) that has the principle part of $E_L/(E_L^2-E_R^2)$, we first integrate over E_L. With the assumption $\Delta_L < \Delta_R$, the integral always passes across the pole at E_R giving

$$I_{2L} = \text{P}\int_{\Delta_L}^{\infty} dE_L \frac{\Delta_L}{\sqrt{E_L^2-\Delta_L^2}} \frac{E_L}{E_L^2-E_R^2} \tag{70}$$

$$= \text{P}\int_{1}^{\infty} dx \frac{1}{\sqrt{x^2-1}} \frac{x}{x^2-y^2} \tag{71}$$

$$= -\frac{1}{\sqrt{y^2-1}} \times \left\{ \begin{array}{ll} \text{arctanh}\sqrt{\frac{y^2-1}{x^2-1}} & (x>y) \\ \text{arctanh}\sqrt{\frac{x^2-1}{y^2-1}} & (x<y) \end{array} \right|_1^{\infty} \tag{72}$$

$$= 0 \quad . \tag{73}$$

We thus find no contribution for f_R in the total integral.

We next compute the E_R integral for the $E_R/(E_L^2 - E_R^2)$ term. We define $w = E_L/\Delta_R$, and note that the integration over E_R depends on whether E_L is greater or less than Δ_R

$$I_{2R} = \text{P}\int_{\Delta_R}^{\infty} dE_R \frac{\Delta_R}{\sqrt{E_R^2-\Delta_R^2}} \frac{E_R}{E_L^2-E_R^2} \tag{74}$$

$$= \text{P}\int_{1}^{\infty} dz \frac{1}{\sqrt{z^2-1}} \frac{z}{z^2-w^2} \tag{75}$$

$$= -\frac{1}{\sqrt{1-w^2}} \times \left\{ \begin{array}{ll} 0 & (w>1) \\ \text{arctan}\sqrt{\frac{x^2-1}{1-w^2}} & (w<1) \end{array} \right|_1^{\infty} \tag{76}$$

$$= -\frac{\pi}{2}\frac{1}{\sqrt{1-w^2}} \; \theta(\Delta_R - E_L) \quad . \tag{77}$$

This result implies that when integrating over E_L, no contribution comes from Δ_R to infinity, so the full integral is

$$I_{2RL} = \int_{\Delta_L}^{\Delta_R} dE_L \frac{\Delta_L}{\sqrt{E_L^2-\Delta_L^2}} \; 2f_L I_{2R} \tag{78}$$

$$= -\,\pi \int_{\Delta_L}^{\Delta_R} f_L \; dE_L \frac{\Delta_L}{\sqrt{E_L^2-\Delta_L^2}} \frac{\Delta_R}{\sqrt{\Delta_R^2-E_L^2}} \quad . \tag{79}$$

In the limit where Δ_L is close to Δ_R such that f_L is constant over the region of integration, the integral can be evaluated

$$I_{2RL} = -\pi f_L(\Delta_L)\; \Delta_L \text{EllipticK}[1-(\Delta_L/\Delta_R)^2] \tag{80}$$

$$\simeq -\frac{\pi^2}{4}\Delta \; 2f_L(\Delta) \quad , \tag{81}$$

where in the last equation we have taken the limit $\Delta_L \to \Delta_R = \Delta$.

For the case of equal gaps Δ, the first two integrals give a Josephson current with a $\sin\delta$ dependence

$$I_{js} = \frac{2}{\pi e R_n}(I_{1LR}+I_{2RL})\sin\delta \tag{82}$$

$$= \frac{\pi}{2}\frac{\Delta}{eR_n}[1-2f_L(\Delta)]\sin\delta \tag{83}$$

$$= \frac{\pi}{2}\frac{\Delta}{eR_n}\tanh[\Delta/2kT]\sin\delta \quad \text{(thermal)} \quad . \tag{84}$$

The last equation assumes a thermal population of quasiparticles given by $f(E) = 1/[1+\exp(E/kT)]$, which yields the Ambegaokar-Baratoff formula [6] for the Josephson current.

The Josephson current can also be calculated for an arbitrary quasiparticle occupation under the assumption that the difference of the gaps $\Delta_R - \Delta_L$ is much larger than the typical width of the quasiparticle distribution. The contribution from the principle part for the f_R term is zero, as discussed before. The contribution from f_L (the lower gap side of the junction) is given by Eq. (79). Noting that f_L is peaked at $E_L = \Delta_L$, we find the current change from quasiparticles is given by

$$I_{j2} \simeq -\frac{2\sin\delta}{eR_n}\frac{\Delta_R}{\sqrt{\Delta_R^2-\Delta_L^2}}\int_{\Delta_L}^{\Delta_R} f_L \; dE_L \frac{\Delta_L}{\sqrt{E_L^2-\Delta_L^2}} \tag{85}$$

$$\simeq -\frac{\sin\delta}{eR_n}\frac{\Delta_R\Delta_L}{\sqrt{\Delta_R^2-\Delta_L^2}} \; \frac{n_{\text{qp}L}}{N_{0L}\Delta_L} \quad , \tag{86}$$

Note that these equations have a contribution from quasiparticle occupation only from the left electrode, which has lower gap. This makes sense since a more exact theory of Andreev bound states has suppression of the critical current from occupied states in the gap, which has to have energy below that of the lowest gap.

For the $\cos\delta$ term, we first note that the current diverges logarithmically for $\Delta_L \to \Delta_R$. Assuming the quaisparticle density is constant with $f = f_l + f_R - f_L f_R$, numerical integration gives the approximate formula

$$I_{jc} \simeq -\frac{2\cos\delta}{eR_n}\big[-0.1+\frac{\Delta_L}{\Delta_R}-0.5\ln\big(1-\frac{\Delta_L}{\Delta_R}\big)\;\big]f \tag{87}$$

For the case where the gaps greatly differ, and in the limit discussed in the previous paragraph, the current is

$$I_{jc} \simeq -\frac{2\cos\delta}{eR_n}\rho_L(\Delta_R)\frac{\Delta_L\Delta_R}{\Delta_R^2}\int_{\Delta_R}^{\infty}\rho_R \, dE \, f_R \tag{88}$$

$$= -\frac{\cos\delta}{eR_n}\frac{\Delta_L^2}{\sqrt{\Delta_R^2-\Delta_L^2}} \; \frac{n_{\text{qp}R}}{N_{0R}\Delta_R} \quad , \tag{89}$$

which has a form and magnitude similar to the thermal current.

Note that for the $\cos\delta$ current, quasiparticles contribute from the higher gap side of the junction. This contrasts the behavior of the $\sin\delta$ current, which has contribution from the superconducting electrode with lower gap.

[1] A. O'Connell, M. Ansmann, R. C. Bialczak, M. Hofheinz, N. Katz, Erik Lucero, C. McKenney, M. Neeley, H. Wang, E. M. Weig, A. N. Cleland, and J. M. Martinis, Appl. Phys. Lett. **92**, 112903 (2008).

[2] D. Mattis and J. Bardeen, Phys. Rev. **111**, 412 (1958).

[3] B Geilikman and V KResin, *Kinetic and Nonsteady-state Effects in Superconductors*, John Wiley and Sons, 1974.

[4] John M. Martinis, M. Ansmann and J. Aumentado, Phys. Rev. Lett. **103**, 097002 (2009).

[5] K. Osbourne and J. Martinis, "Superconducting Qubits and the Physics of Josephson Junctions", Les Houches proceedings and arxiv:cond-mat/0402415 (2004).

[6] V. Ambegaokar and A. Baratoff, "Tunneling Between Superconductors", Phys. Rev. Lett. **10**, 486 (1963).

[7] Note that the integral was checked only for $\Delta_L = \Delta_R$; the general case is taken from Eq. (23) in Ref. [6].