# Cooper-pair tunneling in small junctions with tunable Josephson coupling

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We investigate Cooper-pair tunneling in a circuit consisting of two dc-superconducting quantum interference devices in series, with a gate capacitively coupled to the central island. Measurements cover a wide range of values of the ratio between Josephson coupling energy  $E_J$  and charging energy  $E_C$ . The  $E_J/E_C$  ratio dependence of the supercurrent is well described by the orthodox theory provided that strong fluctuations of the Josephson phase due to the electromagnetic environment are taken into account. Our data can be interpreted in terms of squeezing of the charge fluctuations with decreasing  $E_J/E_C$  ratio. [S0163-1829(96)06533-2]

### I. INTRODUCTION

Nanometer-scale technology enables us to make ultrasmall tunnel junctions with the area less than 0.01  $\mu$ m<sup>2</sup>. In such tunnel junctions the tunneling of electrons is blocked at low temperatures  $k_B T \ll E_C$  and low bias voltages  $eV \ll E_C$ (here  $E_C = e^2/2C$  is the charging energy). This phenomenon is called a Coulomb blockade.<sup>1-3</sup> When the junction becomes superconducting, the Josephson coupling energy  $E_{I}$ must be taken into account. The Josephson coupling energy characterizes the strength of the phase correlation between two superconducting electrodes of the junction, or the wave nature of the pseudowave function of a superconductor. On the contrary, the charging energy characterizes the correlation in the number of particles in the electrodes, or the particle nature of the superconducting wave function. Thus, we can expect a different behavior of the system depending on the ratio between  $E_I$  and  $E_C$ .

In a more formal way, the Josephson phase difference  $\phi$  and the charge Q on the junction are canonically conjugated variables  $[\phi, Q] = 2ie$  satisfying the Heisenberg uncertainty relation  $\Delta \phi \Delta Q > e^{.4}$  With decreasing  $E_J/E_C$  ratio the quantum fluctuations of the phase increase and the fluctuations of the charge decrease. One can say that the charge fluctuations get squeezed.

The competition between the charging energy and the Josephson coupling energy can be investigated using a modified superconducting single-electron transistor (S-SET). <sup>5,6</sup> In this device the effective Josephson coupling energy  $E_J^*$  can be controlled by a small magnetic field. A variation of this method was used in Ref. 7, where the switching current of a controllable double junction was investigated as a function of the gate voltage.

A simple "orthodox" theory for the Josephson current through S-SET was presented in Ref. 1. The modification of the Josephson current due to the parity effects was studied both theoretically<sup>8</sup> and experimentally.<sup>9,10</sup> These studies were concentrated on the case of small Josephson coupling energy  $(E_J \ll E_C)$ . A systematic investigation of the parity effects for an arbitrary  $E_J/E_C$  ratio seems to be an interesting open problem.

Besides the Coulomb and parity effects, the Josephson current is influenced by the dissipation due to the electromagnetic environment.<sup>11–15</sup> Sufficiently strong quantum fluctuations of the environment can delocalize the Josephson phase and suppress the Josephson effect completely.<sup>11</sup> However, typically the quantum fluctuations are weak  $\text{Re}[Z_{\text{env}}]/(h/4e^2) \ll 1$  due to the low impedance  $Z_{\text{env}}$  of the environment. On the other hand, the classical thermal fluctuations of the phase can be substantial due to the coupling of the junction to a high-temperature part of the circuit.<sup>16</sup> These fluctuations should be properly taken into account when one analyzes experimental data.

In this paper, we study the current-voltage (I-V) curves of modified superconducting single-electron transistors (S-SET's) at various magnetic fields and gate voltages. We found that (1) the experimental dependences of the supercurrent on the  $E_J/E_C$  ratio can be explained by the orthodox theory provided that the fluctuations of the Josephson phase due to the electromagnetic environment are taken into account. These fluctuations are characterized by the effective temperature which in our case was substantially higher than the base temperature. (2) The charge fluctuation squeezes as the  $E_J/E_C$  ratio decreases.

In Sec. II we briefly describe the device configuration and its fabrication process. To change an effective Josephson coupling energy in the system, we used a dc-superconducting quantum interference device (SQUID) geometry. Junctions were made of aluminum/aluminum oxide/niobium. In Sec. III we show experimental data that were obtained from I-V curves at various magnetic fields and gate voltages. We found two specific features for supercurrent: unexpected small amplitude and its finite slope. In Sec. IV we assume that these features came from the fluctuation of the Josephson phase due to the external electromagnetic environment and derive the expression for the Josephson current. This analysis shows good agreement with measurements. The uncertainty principle between the phase and charge is also demonstrated. It is surprising to see a squeezing of the charge fluctuation in our samples where the external noise is rather large. In Sec. V we summarize our results.

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FIG. 1. The configuration of a modified superconducting singleelectron transistor (S-SET).

#### **II. FABRICATION AND MEASUREMENT SYSTEM**

We fabricated modified S-SET's whose geometry is composed of two dc-SQUID's in series with a capacitively coupled gate electrode, as shown in Fig. 1. The mean area of the dc-SQUID loop S is  $0.61 \mu m^2$  and the central island is 0.3  $\mu$ m<sup>2</sup>. The junctions are made of Al/AlO<sub>x</sub>/Nb. Since we used different materials for the base and counter electrodes, the barrier layer of  $AlO_x$  covered the edge of the niobium base to prevent current leakage. The current is injected from the Al electrode and goes to the other Al electrode after passing through the Nb central island. A dc-SQUID can be regarded as a single junction with an effective Josephson coupling energy  $E_J^*$ . A dc-SQUID geometry allows us to change the effective Josephson coupling energy  $E_I^*$  when a small external magnetic field is applied. The equivalent circuit of the modified S-SET is shown in Fig. 2, where  $E_{Ix}^*$ (x=1,2) represents the effective Josephson coupling energy of a dc-SQUID. Assuming that each junction of a dc-SQUID is identical, the effective Josephson coupling energy can be expressed as follows,  $E_I^*(f) = 2E_I |\cos(\pi f)|$ , where the frustration is  $f = \Phi/\Phi_0$ ,  $\Phi$  is an applied flux in a SQUID loop and  $\Phi_0 = h/2e \approx 20.7 \text{ G}/\mu\text{m}^2$  is a flux quantum. The sample is symmetrically biased to reduce the noise. Due to the stray capacitance of leads, the sample is always voltage-biased even when an ideal current source is attached.

The S-SET's were fabricated using electron-beam lithography and the shadow mask evaporation technique. The fabrication process is as follows. A clean silicon substrate with



FIG. 2. The equivalent circuit of an S-SET. The dc-SQUID's were replaced by single junctions with an effective Josephson coupling energy.

a 2- $\mu$ m-thick layer of thermal oxided SiO<sub>2</sub> is used for the fabrication. The oxide layer of a Si substrate enables us to measure samples at room temperature. First, the four-layer resist system, which is composed of polymethylmethacrylate (PMMA) as the top layer, germanium, hard-baked photoresist, and a base layer of PMMA, is prepared. A detailed preparation procedure of the four-layer resist is described elsewhere.<sup>6</sup> The top layer of PMMA has a high resolution and the bottom PMMA layer is just used for easy lift-off, because a hard-baked photoresist sticks to the substrate. The advantage of the four-layer resist system compared with the standard double-layer resist<sup>17</sup> is the possibility of optimizing the resolution of the top PMMA. Moreover, the suspended Ge bridge is tough enough to evaporate hard materials, such as niobium.

The base electrodes of 20-nm-thick niobium were deposited on a clean silicon substrate with the thermal oxide layer by electron-gun evaporation at the angle of  $-15^{\circ}$  to the normal of the substrate. Then 7-nm-thick aluminum was evaporated at a slightly different angle to cover the edge of the Nb central island. After the evaporation of aluminum, 20 mTorr oxygen was introduced to form a tunnel barrier, and finally, the counter electrodes of 40-nm-thick aluminum were deposited at the opposite angle,  $15^{\circ}$ . Therefore, the central island was made of niobium and the outer electrodes were aluminum.

Measurements were performed in a top-loading dilution refrigerator whose base temperature was about 20 mK. The cryostat is situated in an electrically shielded room. Samples were symmetrically biased to avoid pickup of external noise, and measurement leads with RC and LC filters at different temperature stages were designed to reduce thermal noise, but we did not use the copper powder filter.<sup>16</sup> A computer-ized data acquisition system was used, which enabled us to take a large number of *I-V* curves continuously, while stepping gate voltage, magnetic field, and temperature.

# **III. RESULTS**

We measured I-V curves of an S-SET at various gate voltages and magnetic fields. Typical *I-V* curves at T=20 mK are shown in Fig. 3. Remarkable current peaks at  $V \approx \pm 670 \ \mu V$  in Fig. 3(a) are the so-called Josephsonquasiparticle (JQP) peaks,<sup>18</sup> where the peak voltage corresponds to  $V \approx 2\Delta + E_C$ . The JQP peak stems from the resonant tunneling of a Cooper pair accompanied by the tunneling of two quasiparticles.<sup>5,6,14,18</sup> The JQP peak, as well as the supercurrent, oscillates with the period of  $29.7\pm0.63$ G when a small magnetic field is swept. This measured period agrees with the calculated one  $\Delta B = \Phi_0 / S = 34$  G, where S is the mean area of the SQUID loop. The supercurrent is modulated from  $I_{C \text{ max}} = 2.54$  nA to  $I_{C \text{min}} = 18.2$  pA at the fixed gate voltage  $V_g = 0$  by applied magnetic field and thus the modulation ratio of supercurrent is  $\gamma = I_{Cmax}/I_{Cmin} = 140$ . This high value of  $\gamma$  indicates that the junctions are almost identical.<sup>19</sup> The normal resistance and the capacitance of a junction can be estimated as  $R_N = 11.4$  $k\Omega$  and C=0.28 fF, where the junction capacitance was estimated from its geometry. The I-V characteristics were also modulated by the gate voltage with the period of  $\Delta V_{g} = 11.3$  mV, and thus the gate capacitance is



FIG. 3. Typical *I-V* curves at T=20 mK; (a) at middle scale, (b) at vicinity of the origin, where the solid line and broken line were obtained at the gate voltage  $V_g=0$  and  $V_g=5.7 \mu$ V, and (c) at large scale.

 $C_g = e/\Delta V_g = 14.2$  aF. Therefore, the charging energy can be estimated as  $E_C = e^2/2C_{\Sigma} \approx 70 \ \mu eV$ , where  $C_{\Sigma} = 4C + C_g$  is the total capacitance of the central island.

The *I-V* curves at low bias voltage are shown in Fig. 3(b). The supercurrent has the slope and its peak is located at about  $V=35 \ \mu$ V. The current modulation was *e* periodic in the gate-induced charge  $Q_g = C_g V_g$ . Although *e* periodicity does not agree with the parity theory<sup>8</sup> it was observed in many experiments<sup>3</sup> (we will comment later on that). The current peaks observed at certain bias and gate voltages [see, e.g., a peak  $V=150 \ \mu$ V in Fig. 3(b)] stem from the resonant tunneling of Cooper pairs.<sup>5,6,14</sup>

The large scale of the *I-V* curve is shown in Fig. 3(c), where the supercurrent is suppressed by a small magnetic field. As junctions are composed of different materials, the *I-V* curve shows the relatively broad transition at  $V = \pm 1.2$ mV. Superconducting energy gaps  $\Delta_{\rm Nb} \approx 455 \ \mu eV$  and  $\Delta_{\rm Al} \approx 185 \ \mu eV$  were derived from the derivative of the *I-V* 



FIG. 4. Three-dimensional plot of the critical current for an S-SET as a function of both magnetic field and gate voltage at T=30 mK. X and Y axes are converted into the units of flux  $\Phi=BS$  and gate-induced charge  $Q=C_gV_g$  which are normalized by units of flux quantum  $\Phi_0$  and elementary charge e.

curve, which might have an uncertainty due to the charging effect. However, the error should be small compared with energy gaps and thus we neglect it. The relatively small value of  $\Delta_{\rm Nb}$  is due to the small thickness of the Nb film and the relatively high background pressure during the evaporation also contributed to the smaller energy gap. The Josephson coupling energy was then deduced to be  $E_J = (R_Q/R_N)(\Delta_{\rm Al} + \Delta_{\rm Nb})/4 \approx 90 \ \mu {\rm eV}$  in zero magnetic field, so that  $E_J^*(0)/E_C = 2.6$ , where  $R_Q = h/4e^2 \approx 6.45 \ {\rm k}\Omega$ , and  $R_N$  is the normal resistance of the junction.

Both the magnetic field and the gate voltage dependence of the critical current are plotted in Fig. 4. The critical current is defined as the maximum supercurrent peak of the *I-V* curve, which is located at the bias voltage  $V_p \sim 35 \ \mu$ V. The finite slope of the supercurrent or zero-bias conductance  $G_0$ is always observed. The zero-bias conductance  $G_0$  oscillates periodically as the  $E_J/E_C$  ratio is varied, as shown in Fig. 5. Note that the zero-bias resistance is always larger than  $10^4 \ \Omega$ .

#### **IV. DISCUSSION**

As shown in the previous section, the supercurrent has a finite slope. In addition, the fact that the measured maximum



FIG. 5. The zero-bias conductance  $G_0$  as a function of frustration. The solid, broken, and dotted lines are taken at the temperature T=1.0, 0.6, and 0.03 K.

supercurrent  $I_c^{\text{max}} \simeq 2.54$  nA through the S-SET is much less than the maximum supercurrent estimated by means of the Ambegaokar-Baratof formula,  $I_c^{\text{est}} = 4eE_J/\hbar \simeq 90$  nA implies a strong effect of the phase fluctuations. The source of these fluctuations could be a high-frequency noise from heliumtemperature or a higher-temperature part of the measurement circuit (electromagnetic environment), which can be appreciable even after strong attenuation.<sup>16</sup>

Since no resistors are placed in the vicinity of the junction, its electromagnetic environment can be modeled by an *LC* line with Ohmic impedance of the order of free space impedance,  $Z_{\rm env} \sim 100 \ \Omega$  (see, e.g., Refs. 3,16). As this value is much lower than the quantum resistance  $h/e^2 \sim 25.8 \ k\Omega$ , one can neglect the quantum fluctuation and consider only thermal fluctuation of electromagnetic quantities. We will characterize this fluctuation by effective temperature of the environment,  $T_{\rm env}$ .

As a first step, we compute the Josephson current  $I_J^{(0)}$  as a function of the phase difference  $\phi = \phi_l - \phi_r$ , which is assumed to be fixed.<sup>8</sup> The Hamiltonian of the system in the charge representation has the form

$$H = E_C(n) |n\rangle \langle n| - E_J\{(e^{i\phi_l} + e^{i\phi_r}) |n+1\rangle \langle n| + \text{H.c.}\}, \qquad (1)$$

where  $E_C(n) = (2en - Q_g)^2/2C_{\Sigma}$  is the Coulomb energy of the state with *n* Cooper pairs on the central electrode. Since the spectrum is 2*e* periodic in the gate charge  $Q_g$ , it is enough to consider the interval  $-e < Q_g < e$ . The Josephson current can be found as a derivative of the free energy *F* with respect to the phase,

$$I_J(\phi) = -2e \,\partial F / \partial \phi. \tag{2}$$

We calculate the free energy  $F = -\ln \sum_p \sum_i \exp(-E_{p,i}/k_BT)$  by computing the eigenvalues  $E_{p,i}$  of the Hamiltonian (1) numerically. Here  $\sum_p$  denotes the summation over the states with even (p=0) and odd (p=1) numbers of electrons on the island.<sup>9,10</sup> Since we have no signatures of 2*e*-periodic behavior of the Josephson current as a function of gate voltage in the experimental data, we assume that there is a number of quasiparticle states in the superconducting gap. For this reason, we do *not* introduce the parity-dependent term (related to a finite value of the superconducting gap) into the energies of states with an odd number of electrons (p=1). Hence,  $E_{1,i}$  simply correspond to the eigenvalues of the Hamiltonian (1) with shifted  $Q_g$ ,  $Q_g \rightarrow Q_g - e \operatorname{sign} Q_g$ , whereas  $E_{0,i}$  correspond to the eigenvalues without this shift. In this case *e* periodicity is fulfilled automatically.

The critical current is defined as  $I_{c,0} \equiv 2eE_{J,eff}/\hbar = \max_{\phi} |I_J(\phi)|$ . In particular, for  $E_J, k_B T \ll E_C$  we obtain

$$E_{J,\text{eff}} = (E_J^2/2) [1/E_1 + 1/E_2], \qquad (3)$$

where  $E_{1(2)} = E_C(\pm 2) - E_C(0)$  are the Coulomb energies of two intermediate states for the tunneling of a Cooper pair through the transistor.

The next step is to take into account the fluctuations. We will model the fluctuations of the voltage on the transistor by a white noise whose intensity is proportional to the Ohmic impedance  $Z_{env}$  of the environment,

$$\langle V_{\xi}(t)V_{\xi}(t')\rangle = 2Z_{\rm env}k_B T_{\rm env}\delta(t-t'). \tag{4}$$

Note that the temperature of the electromagnetic environment  $T_{env}$  is typically larger than the actual temperature T of the transistor due to the spurious influence of high-frequency components of external noise.<sup>16</sup>

Since  $\dot{\phi} = 2eV/\hbar$ , then phase difference  $\phi$  will show diffusive behavior,  $\langle \phi^2(t) \rangle = 4Dt$  (in the absence of the Josephson potential) with  $D = (2e/\hbar)^2 Z_{env} k_B T_{env}$ . Generally, the effect of thermal fluctuation is described by the Smoluchovski equation for the distribution function  $\sigma(\phi)$  of the phase, <sup>20</sup>

$$\frac{\partial \sigma}{\partial t} + \frac{2eZ_{\rm env}}{\hbar} \frac{\partial}{\partial \phi} \{ [I_{\rm ex} - I_J(\phi)]\sigma \} = \frac{(2e)^2}{\hbar^2} Z_{\rm env} k_B T_{\rm env} \frac{\partial^2 \sigma}{\partial \phi^2},$$
(5)

where  $I_{ex}$  is the external current and  $I_J(\phi)$  is given by Eq. (2). The stationary solution  $\sigma(\phi)$  of this equation enables one to find the average Josephson current through the transistor,  $\overline{I}_J = \int_{-\pi}^{\pi} d\phi \sigma(\phi) I_J(\phi)$  and the voltage on it,  $V = Z_{env}(I_{ex} - \overline{I}_J)$ . Note that the impedance of the environment does not enter into the *stationary* Smoluchovski equation. Hence, the maximum Josephson current in presence of fluctuations  $I_{c, fluct} = \max_{I_{ex}} |\overline{I}_J|$  does not depend on the impedance of the environment.

In the limit of strong thermal fluctuations,  $k_B T_{env} \gg E_{J,eff}$ we obtain the *I-V* characteristic,

$$\overline{I}_{J} \simeq \frac{Z_{\text{env}} I_{c,0}^2}{2} \frac{V}{V^2 + V_p^2} \tag{6}$$

with  $V_p = (2e/\hbar)Z_{env}k_BT_{env}$  [exact equality holds for harmonic dependence  $I_J(\phi)$ ]. This determines the maximum Josephson current  $I_{c,fluct} \approx (2e/\hbar)E_{J,eff}^2/k_BT_{env}$  and the zerobias conductance  $G_0 \approx (E_{J,eff}/k_BT_{env})^2/2Z_{env}$ . From the experimental *I-V* curves we obtain

From the experimental *I-V* curves we obtain  $V_p = 35 \ \mu V$  which gives  $k_B T_{env} \approx 360 \ \mu eV \ (T_{env} \approx 4.2 \ K)$  for  $Z_{env} = 200 \ \Omega$ .  $Z_{env}$  was used for the measured resistance of the lead. Using this estimate and the maximum zero-bias conductance  $G_0 = 1.2 \times 10^{-4} \ \Omega^{-1}$  we obtain  $E_{J,eff} \approx 80 \ \mu eV$ , which is in agreement with the calculated value  $E_{J,eff} = 90 \ \mu eV$  for f = 0.

Let us now return to Fig. 5 and concentrate on the dependence of zero-bias conductance  $G_0$  on temperature. As the temperature is increased,  $G_0$  decreases slightly at f=0 and 1. Indeed, the temperature of the environment is much higher than the system temperature T. For this reason, an increase in T changes  $T_{env}$  and decreases  $G_0$  only slightly. On the other hand,  $G_0$  appreciably *increases* with temperature for f=0.5and 1.5. The Josephson tunneling is strongly suppressed in this regime  $[I_{c,\text{fluct}} \propto E_{L,\text{eff}}^2 \propto (f-1/2)^4]$ . The increase of  $G_0$ with temperature can be explained by thermally activated transport of quasiparticles. In fact, the anomalous temperature dependence of the critical current was observed (see the inset of Fig. 6). The enhancement of the critical current at  $T \sim 900$  mK may imply the existence of excess quasiparticles in the central island according to the Eliashberg mechanism in a nonequilibrium superconductor.<sup>21</sup> We do not have any idea to explain the peak at T=0.2 K. The study of the zerobias resistance at various values of the  $E_I/E_C$  ratio and the external impedance is a further subject.



FIG. 6. The  $E_J/E_C$  ratio dependence of the critical current at T=30 mK. The gate voltage is fixed at  $V_g=0$ . The solid and broken lines are calculated by the orthodox theory with and without thermal fluctuations ( $T_{\rm env}=4.2$  K) of Josephson phase, where  $E_J^*(0)/E_C=2.6$  is assumed. Measured data are shown by circles. The inset shows the temperature dependence of the critical current at f=0.

The dependence of the critical current on the frustration (or  $E_J^*/E_C$ ) is shown in Fig. 6. The measured data are represented by circles. The current is normalized by the maximum current value. The broken line shows the critical current  $I_{c,0}$  in the absence of fluctuations of the Josephson phase (for  $E_J^*(0)/E_C=2.6$ ). The solid line corresponds to the result with fluctuations ( $I_{c,fluct}$ ) which is roughly proportional to  $I_{c,0}^2$  since the condition  $k_B T_{env} \ge E_{J,eff}$  is fulfilled. This curve corresponds much better to the experimental data.

The modulation depth  $(I_{C \max} - I_{C \min})/I_{C \max}$  of the critical current by the gate voltage is plotted in Fig. 7. The solid and broken curves correspond to the results with and without the fluctuations for  $E_I^*(0)/E_C = 2.6$ . The amplitude of the modulation depth increases as f increases from zero toward 0.5. This is because the charge sensitivity by gate voltage is enhanced at the weak coupling limit where the charge fluctuation is squeezed due to the uncertainty relation.<sup>7</sup> This agrees with the fact that the amplitude of the supercurrent decreases with the smaller  $E_J/E_C$  ratio as shown in Fig. 6, where the phase fluctuations increased as the  $E_J/E_C$  ratio decreased. It might seem surprising that even near f = 0.5 the modulation is still not very strong. An explanation for this is related to the *e* periodicity of experimental data with respect to the gate voltage. Indeed, if one restricts the gate charge  $Q_g$  by the interval [-e/2, e/2], and uses expression (3) for  $E_{J,eff}$ , one obtains that the ratio of the maximum critical current [at  $Q_g = e/2, E_1 = (3e/2)^2/2C_{\Sigma}, E_2 = (5e/2)^2/2C_{\Sigma}$ ] to the minimum critical current [at  $Q_g = 0, E_1 = E_2 = (2e)^2/2C_{\Sigma}$ ] is



FIG. 7. The critical current modulation depth as a function of the  $E_J/E_C$  ratio at T = 30 mK. The modulation depth is defined as  $(I_{Cmax} - I_{Cmin})/I_{Cmax}$ . The solid and broken lines are calculated by the orthodox theory with and without thermal fluctuations  $(T_{env} = 4.2 \text{ K})$  of Josephson phase. Circles represent measured data.

equal to 4/3. Since the critical current with fluctuations is proportional to  $E_{J,\text{eff}}^2$  the ratio of the maximum to the minimum critical current will be 16/9, which corresponds to a modulation depth of 43%. The experimentally observed maximum modulation depth (35%) is close to this estimate.

### V. SUMMARY

In summary, we have investigated current-voltage characteristics of modified superconducting single-electron transistors at various magnetic fields and gate voltages. We found that the  $E_J/E_C$  ratio dependence of the supercurrent is well described by the orthodox theory with thermal fluctuations of the Josephson phase due to the electromagnetic environment. The charge fluctuation squeezes as the  $E_J/E_C$  ratio decreases. This agrees with the uncertainty principle. Observed *e*-periodic oscillations in gate charge are attributed to the subgap states in the superconducting energy gap. Experimental and theoretical investigation of the parity effects in the strong coupling limit presents an interesting problem for the future.

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