Charging Energy and Phase Delocalization in Single Very Small Josephson Tunnel Junctions

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We have measured current-voltage characteristics of very small (0.4–0.02 μm^2) Sn-SnO_x-Sn tunnel junctions, having estimated a charging energy $e^2/2C$ comparable to their other characteristic energies. In the higher-R_n devices, after rising just below T_c, I_c *decreases* as the temperature is decreased further, and then *increases* again, at the lowest temperatures. Although the junctions are hysteretic, a significant resistance is found at currents below I_c. We suggest an interpretation involving the quantum nature of ϕ and the competition between the charging, Josephson, and thermal energies of the system.

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The Josephson tunnel junction is well suited for studying macroscopic quantum mechanics. Recent experiments[1-3] show that quantum tunneling of the macroscopic variable ϕ (the phase difference between the junction electrodes) is important at low temperatures, confirming theoretical predictions.[4,5] Our experiments carry this further, to junctions so small that the quantum phase-number uncertainty relation appears to play a major role. This arises since ϕ and Q (the Cooper-pair charge difference between the electrodes) are quantum-mechanical operators with commutator $[\phi,Q]=2ie$. The behavior of the device is determined by the ratio of two energies: $U_J=-E_J\cos\phi$ associated with ϕ, and $U_c=(Q^2/e^2)E_c\rightarrow -4E_c\,\partial^2/\partial\phi^2$ associated with Q. Here $E_J=\hbar I_{c0}/(2e)$ is the Josephson energy, $E_c=e^2/2C$ is the charging energy. Furthermore, $I_{c0}=(\pi\Delta/2eR_n)\times\tanh(\Delta/2k_BT)$ is the unfluctuated critical current given by Ambegaokar and Baratoff,[6] C is the capacitance, Δ is the superconducting energy gap, and R_n is the normal resistance.

We have fabricated very small tunnel junctions with estimated $E_c \gtrsim E_J$; their behavior is fundamentally different from that of conventional "semiclassical" junctions, with $E_J \gg E_c$. Figure 1 shows the $I_c(T)$ for all samples measured. As R_n increases from 520 Ω for $S1$ to 70 kΩ for $S7$, we see a dramatic change in behavior. While $I_c(T)$ for the low-resistance samples displays the usual monotonic increase with decreasing temperature, the behavior of the high-R_n samples is strikingly reentrant, I_c *decreasing* with decreasing T at intermediate temperatures and *increasing* again at low temperatures. Furthermore, the I-V curves of the high-resistance samples exhibit an anomalous resistance R_0 below I_c, while being strongly hysteretic. While the high-temperature data may be understood in terms of classical effects in an unusual regime, a full quantum-mechanical treatment seems necessary to interpret the low-temperature results.

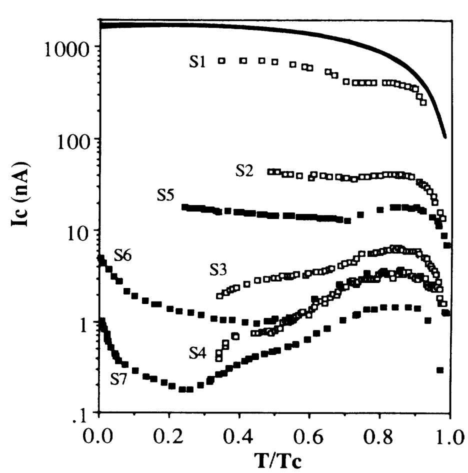

FIG. 1. I_c vs T for all samples. The solid curve is $I_{c0}(T)$ for $S1$.

The Sn-Sn junctions were patterned by a two-layer electron-beam lithography technique[7] and completed in one vacuum run. The electrodes were evaporated onto the liquid-nitrogen–cooled substrate and the oxide barrier was grown by a glow discharge. The low dielectric constant of SnO_x, the small width (0.2–0.4 μm) of the in-line electrodes, and the absence of nearby ground planes reduce the intrinsic capacitance of the device. The use of nonsuperconducting leads (up until $\simeq 30$ μm from the device) and the existence of a second slightly larger junction in series with the device studied (except for $S2$ and $S6$) may have helped reduce the effective parasitic capacitance added by the external circuit. Granular structure in the Sn film, such as may have affected our earlier observations,[8] was avoided. Sample parameters are listed in Table I. R_L is the subgap leakage resistance, defined as the slope of the linear part of the sharply dropping branch of the I-V curve; typically, $R_L^{-1}\approx R_n^{-1}\exp(-\Delta/k_BT)+R_{L0}^{-1}$, with $R_{L0}\sim 100R_n$. Capacitances are estimated[8] from measurements of the junction area. All samples had critical temperatures $T_c \approx 3.75$ K.

Samples $S1$–$S4$ were run in a ^{4}He cryostat; $S5$–$S7$, in a dilution refrigerator. Both setups were in screened rooms, and effort was spent to avoid extraneous electrical

TABLE I. Sample parameters; definitions of R_n, I_{c0} (here evaluated at $T=0$), R_L (measured at $T=T_{\min}$), C, E_J, E_c, and I_c^Z are given in the text. $T_{\min}$ is the lowest temperature at which the sample was measured. $I_c^* = I_c(T=23\text{ mK})$.

Sample	R_n (kΩ)	R_L (kΩ)	$T_{\min}$ (K)	Area [(μm)²]	C (fF)	E_J (K)	E_c (K)	I_{c0} (nA)	I_c^Z (nA)	I_c^* (nA)
S1	0.52	10	1.4	0.12	2	40	0.45	1810		
S2	3.4	105	1.8	0.16	3	6.2	0.3	277		
S3	30	2500	1.3	0.023	1	0.7	0.9	31		
S4	40	6300	1.3	...	...	0.5	...	23		
S5	6.5	3000	0.85	0.4	7	3.2	0.13	145		
S6	34	2400	0.023	0.12	2	0.6	0.45	28	7	5.1
S7	70	40000	0.023	0.03	1	0.3	0.9	14	0.9	1.15

pickup and to ensure that the samples were well heat sunk. Figure 2 shows the high quality I-V characteristics of $S7$, at $T=0.98$ K, and definitions of I_c and I_r. Plots of the reentrant I_c and I_r vs T for $S7$ are shown in Fig. 3(a). In the hysteretic regime ($T<0.58T_c$), the increase in voltage at $I=I_c$ was sharp, as shown in Fig. 2, and the distribution of switching currents was narrow, of width less than $0.05I_c$. The presence of a resistive voltage at all currents [see Fig. 2(a)] is common to the higher-R_n samples; we characterize it by the resistance $R_0=dV/dI$ as $I\rightarrow 0$. Figure 4 shows the temperature dependence of R_0. While in low-R_n samples, where R_0 decreases rapidly with temperature, becoming immeasurable soon after hysteresis sets in, high-R_n samples ($R_n > R_Q = h/4e^2 \sim 6.5$ kΩ) exhibit a significant R_0 at all temperatures.

Extensive work has been done[9,10] on the I_c depression in a "classical" ($E_J \gg E_c$) underdamped Josephson junction due to thermally activated premature switching from the zero-voltage state to the gap-voltage state. (Our samples were underdamped, typically having $\beta_c = 2eI_{c0}R_L^2C/\hbar > 5000$.) For $S1$, the average I_c reduction and width of the distribution (at $T \simeq 1.5$ K) are consistent with predictions from this theory. For the other samples, however, the predicted escape rate is much larger than the sweep rate, even at $I=0$, since E_J is not large as compared with k_BT (except as $T\rightarrow 0$): ϕ should constantly be activated out of the Josephson potential well and keep increasing, provided that the energy gained from the bias current exceeds that lost by damping. This is the condition determining I_r, so one would expect $I_c = I_r$, with no I-V curve hysteresis. The predicted[11] $I_r = (4/\pi)\{I_{c0}(T)/[B_c(t)]^{1/2}\}$, in the absence of thermal fluctuations, is shown in Fig. 3(b), with the assumption that $R_L \sim R_n \exp(\Delta/k_BT)$ is the source of damping, and that $C=1$ fF. While the shape of I_r vs T is well explained by this model, its predicted magnitude is smaller than is measured. However, analytical approximations[12] show that thermal fluctuations may greatly increase the predicted I_r. Our digital simulations (which include damping as a piecewise linear resistance) show that the discrepancy in magnitude can be accounted for, within a factor of 2, by including thermal noise, and confirm that $I_c = I_r$. Thus, in the nonhysteretic regime,

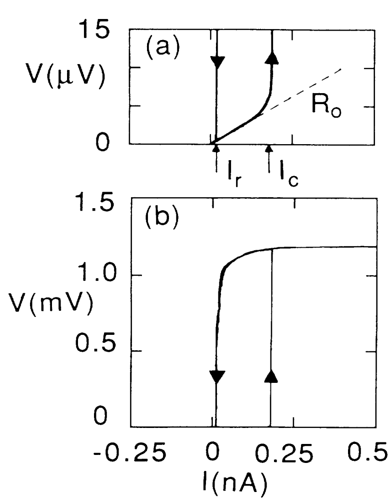

FIG. 2. I-V curve of sample $S7$ at $T=0.98$ K, showing definitions of I_c, I_r, and R_0. Parts (a) and (b) have the same horizontal scale but different vertical scales.

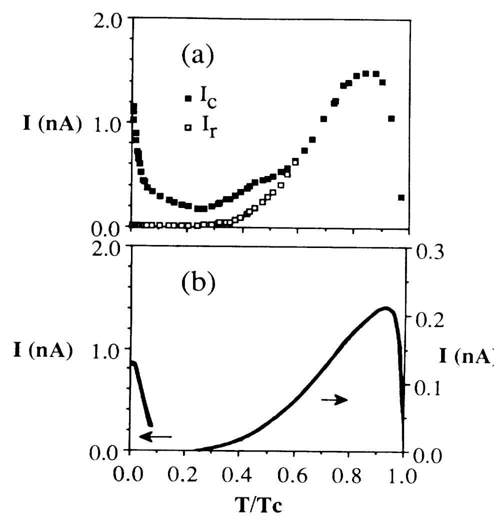

FIG. 3. (a) I_c and I_r vs T for sample $S7$. (b) On the left is the predicted low-temperature I_c, due to Zener tunneling and thermal activation. On the right is the predicted $I_r(T)$ as described in the text.

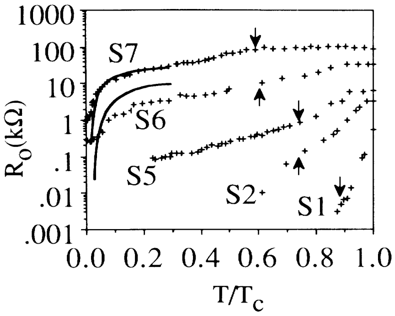

FIG. 4. R_0 vs T for five samples. Below $T \sim 0.85T_c$ for $S1$ and $0.60T_c$ for $S2$, R_0 was below our experimental resolution. The arrows indicate the temperatures below which the I-V curves are hysteretic. The solid lines are theoretical fits for $S6$ and $S7$ as described in the text.

the exponential freeze-out of damping causes the reentrant drop in I_c. At lower temperatures, however, this model breaks down.

Our low-temperature data display two main puzzling features. First, the I-V curves become hysteretic but still display a sharply defined I_c with a narrow distribution. The measured I_c, however, even if extrapolated to $T=0$ (as can be done with some confidence with samples $S6$ and $S7$) is an order-of-magnitude below the unfluctuated value I_{c0}. Second, all high-resistance junctions show an anomalous resistance R_0. These features appear incompatible with classical models of junction dynamics with predict $I_c = I_{c0}$ at $T=0$, even if the damping is considered to be frequency dependent, as in the work of Ono *et al.*[13] They point out that an implausible temperature of 1 K is needed in a classical fit to the I-V curve of their $R_n = 50$ kΩ junction, taken nominally at 10 mK. Since classical arguments appear to fail, we attempt to explain our data using a quantum-mechanical model. Since the estimated charging energy is large, and $I_c \ll I_{c0}$, standard quantum tunneling models based on a cubic approximation to the Josephson potential[4,5] seem inappropriate. In the absence of a complete theory to describe a Josephson junction in the limit of $E_J \sim E_c$, we offer a simple semiquantitative interpretation of our results. While we neglect the frequency dependence of the damping and possible parasitic contributions of the leads, our approach allows us to probe the consequences of large quantum phase uncertainty and significant energy level width, which arise in the small capacitance limit. We leave a more complete treatment to future work.

The phase is described by a wave function $\psi(\phi)$. If $E_c \gtrsim E_J \gg k_B T$, $\hbar/RC$, as is appropriate for $S6$ and especially $S7$ at low temperatures, $\psi(\phi)$ is not sharply localized within a single potential well, but may be extended in ϕ space and described by Bloch functions[14,15] $\psi_q^s = u_q^s(\phi)\exp(iq\phi/2e)$. The energy spectrum of the system has a band structure: s is the band index, and q is the "quasicharge," analogous to the crystal momentum in a solid. Bandwidths scale with E_c, and increase with s, while the band gaps scale with E_J, and decrease as s increases. In this limit, for *very* small currents, Likharev and Zorin[14] predict static solutions with $q=q_0$ and $V=IR_L$ until, at $I \sim I_t \sim e/(R_L C)$ ($\simeq$5–30 pA, for $S6$ and $S7$), the voltage sharply decreases. We find no evidence of this voltage spike. For $I > I_t$, increasing solutions $q=q(t)$ are expected. These result in oscillations in Q (and V) analogous to Bloch oscillations in a periodic conductor under a large electric field, with frequency $\omega_B = (\pi/e)(I - \bar{V}/R_L)$, where $\bar{V}/R_L$ is typically very small.

We expect that interband transitions, which may occur by a tunneling process analogous to Zener tunneling in solids,[14,16] affect the I-V response of the device. The tunneling rate is[16]

$$\tau_Z^{-1} \sim f_A \exp[-(\pi^2/8)E_J^2/(E_c \hbar\omega_B)], \quad (1)$$

where $f_A \sim \omega_B/2\pi$ is the attempt frequency. For $T>0$, interband transitions will also be induced by thermal activation at an estimated rate $\tau_{\text{th}}^{-1} \sim f_A \exp(-E_J/k_B T)$. After being excited to a higher band, the system will tend to relax back down to the lowest band, discharging the quantum capacitor composed of the junction electrodes. For $\tau_Z^{-1} \ll f_A$, interband transitions are rare but will cause a small dissipative voltage. This voltage increases with I (since $\omega_B \sim \pi I/e$) until one reaches $\tau_Z^{-1} \sim f_A$. Here, Zener tunneling is so common that the band gaps are no longer effective; as a result the capacitor is unable to discharge Cooper pairs at a rate to keep up with the bias current. The band model then breaks down and the voltage rapidly rises to $2\Delta/e$. We associate this "Zener breakdown" with I_c and define I_c^Z by the condition $\tau_Z^{-1} \sim f_A e^{-1}$, obtaining

$$I_c^Z \sim (\pi e/8\hbar)(E_J^2/E_c). \quad (2)$$

[A similar I_c estimate $(eE_J^2/4\hbar E_c)$ is found by use of a perturbation calculation to estimate the energy lowering of the extended ground state $\psi_0(\phi)$ because of the potential $-E_J\cos\phi$.] The critical current of the junction, then, does not appear to be determined by a stochastic activation or tunneling process, but by a limitation of its ability to carry a supercurrent. This accounts for the observed narrow ($\simeq$5%) distribution of I_c values, even when $I_c \ll I_{c0}$. In Table I, values of I_c^Z calculated with use of (2) are compared with measured values of I_c at 23 mK, for $S6$ and $S7$. The agreement appears quite good to us, considering the roughness of the argument and the uncertainty in C. We obtain a temperature-dependent I_c, shown in Fig. 3(b), by setting $\tau_Z^{-1} + \tau_{\text{th}}^{-1} \sim f_A e^{-1}$, generalizing the argument leading to (2). This last estimate should only serve as a rough guide, since we expect this simple additive rate approximation to be oversimplified, in analogy with macroscopic quantum tunneling calculations.[5]

For $I \ll I_c$, infrequent band transitions generate a voltage of order e/C relaxed by damping in time RC. To estimate R_0 phenomenologically, we write $V \sim (e/C)RC(\tau_Z^{-1}+\tau_{\rm th}^{-1})$ ($\simeq IR_0$, for small I). The data are fitted better with use of $R=R_n$, instead of $R=R_L$. The result is shown in Fig. 4. The agreement between our estimate and the data is reasonably good, especially for the smaller capacitance $S7$, for which our model is best suited. We believe the discrepancy observed for $T<80$ mK to be due to the crudeness of our model, although we cannot exclude the possibility of a small amount of extrinsic noise or of an imperfect heat sinking of our sample having affected our lowest temperature measurements.

In conclusion, we present the following picture: At high temperatures, our high-R_n junctions may be described classically, with $I_c=I_r$. As temperature is decreased further, thermal fluctuations are no longer strong enough to dominate quantum tunneling. The I-V curve then becomes hysteretic, keeping $R_0>0$, since the phase can tunnel from one potential well into another without acquiring enough energy for a full escape. When tunneling is important, $\psi(\phi)$ spreads out, becoming less tightly bound by $U(\phi)$, causing a decreased $I_c<I_{c0}$. At the lowest temperatures and damping, $\psi(\phi)$ appears extended and the energy-band picture seems appropriate. The characteristic normal resistance where conventional predictions break down appears to be of the order of the "quantum" resistance R_Q. At this point, using the Ambegaokar-Baratoff formula,[6] we have $E_c \sim E_J \sim k_B T$, for $T\sim 1$ K and $C \sim 1$ fF. As E_c becomes important, the quantum uncertainty in the phase must increase, and a Bloch-function expansion for $\psi(\phi)$ is indicated at low temperatures.

Other authors[17,18] predict that the junction resistance will directly affect the nature of $\psi(\phi)$. At $T=0$, for resistances $R<R_Q$, ϕ is localized; dissipation suppresses quantum tunneling. For $R>R_Q$, ϕ is extended. These predictions have been used[18] to interpret data[19] on granular films. While these models might form a basis to estimate our I_c, it is not clear which value of R should be used. While our data show that R_L is appropriate in the classical argument determining I_r, the low-temperature R_0 data seem better fitted with a relaxation time $\simeq R_nC$, in agreement with the evidence of Washburn *et al.*[2] that a resistance of order R_n is relevant for tunneling.

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