Crossover from Josephson Tunneling to the Coulomb Blockade in Small Tunnel Junctions

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(Received 17 February 1988)

We have fabricated small tunnel junctions with a variety of ratios of charging energy E_c to Josephson coupling energy E_J and measured their *I-V* characteristics. For $E_c \sim E_J$, the *I-V* curve is resistive at all currents and the critical current is greatly reduced, in agreement with our theoretical estimates. As E_J is further reduced by application of a magnetic field, we find a novel regime in which aspects of the Coulomb blockade of tunneling *coexist* with features typical of Josephson tunneling. At still higher magnetic fields, the junction becomes normal and shows the simple Coulomb blockade.

PACS numbers: 74.50.+r, 05.30.−d, 73.40.Gk

In recent years, new lithography and low-temperature techniques have allowed the fabrication and measurement of devices whose behavior is affected by the capacitive charging energy of a single electron.[1-3] While earlier work centered on granular films, in which large random arrays of low-capacitance tunnel junctions may be formed,[4-8] the new single-junction devices allows us to study a variety of phenomena relating to the quantum mechanics of mesoscopic systems, while avoiding the complications of randomness and collective action.

Here we report measurements of the current-voltage (*I-V*) characteristics of small Sn-SnO_x-Sn tunnel junctions, as we vary the ratio of the charging energy ($E_c = e^2/2C$) to the Josephson coupling energy E_J. By varying E_c/E_J we have swept from the conventional Josephson-effect regime ($E_c \ll E_J$) well into the opposite regime, in which $E_J \ll E_c$, and the behavior is dictated by the charging energy of a single electron. We focus on the crossover between these two extremes.

As reported earlier,[2] we find that as E_J becomes of order E_c, the critical current I_c is greatly reduced, and the *I-V* curve becomes resistive, even at very low bias currents. If we now reduce the Josephson coupling still further by applying a magnetic field, we observe the transition into a novel regime, illustrated by the *I-V* curve shown in Fig. 1. This *I-V* response is suggestive of the Coulomb-blockade effect, which has recently received theoretical attention,[9,10] and has been observed[1,3] in tunnel junctions in which the charging energy is completely dominant. At low voltages, charge is trapped on the electrodes, and the dynamic resistance is very high. For $V > e/2C$, however, the electron can acquire enough energy from the source to make tunneling energetically favorable: The dynamic resistance decreases, causing a knee in the *I-V* curve. The striking new feature of our measurements, in which *both* E_J and E_c are significant, is the *coexistence* of a plateau beginning at $V_b \cong e/2C$, reminiscent of the Coulomb blockade, with other features common to Josephson tunneling, such as a sharp jump from the plateau voltage to the superconducting energy-gap voltage at a "critical current" I_c.

The junctions were patterned by electron-beam lithography and thermally evaporated on a liquid-nitrogen-cooled substrate.[2] The junctions had areas ranging from 0.02 to 0.4 μm^2, normal resistance $R_n \sim 500\ \Omega - 140\ k\Omega$, and estimated capacitance $C \sim 1$-4 fF. Devices were patterned as single junctions, double junctions, and eleven-junction linear arrays. The *I-V* characteristics of the samples were measured in a top-loading dilution refrigerator as a function of magnetic field ($0 < H < 2$ T) and temperature (20 mK $< T < T_c \simeq 3.8$ K). All devices had excellent quasiparticle *I-V* characteristics with leakage resistance $R_L \sim (10^2-10^4)R_n$ at $T \ll T_c$.

Josephson-junction dynamics can be described with use of the macroscopic variable ϕ, the phase difference between junction electrodes, and its quantum mechanical conjugate Q, the charge difference. In conventional Josephson devices, E_J, the energy associated with ϕ, is much larger than the charging energy E_c associated with Q, and ϕ can be treated as a classical variable. As E_c/E_J grows, the quantum uncertainty in ϕ increases, the probability of quantum tunneling of the phase becomes ap-

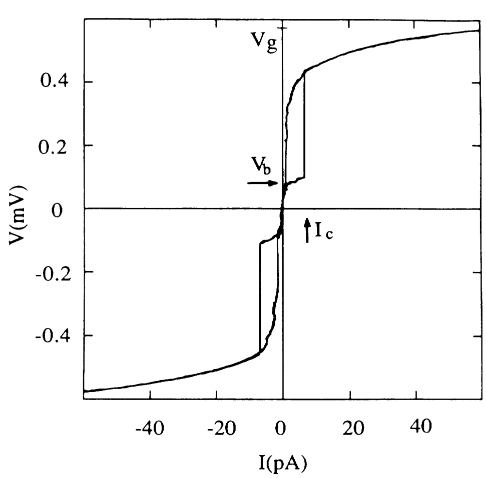

FIG. 1. *I-V* characteristic of a single Josephson junction with $R_n = 140$ kΩ and estimated capacitance $C \simeq 1$ fF in a magnetic field of 0.2 T at $T = 30$ mK.

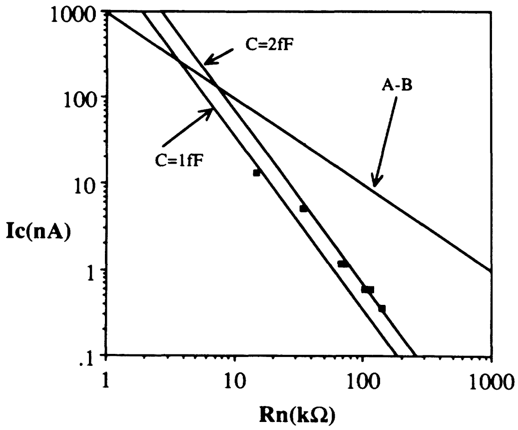

FIG. 2. Measured critical current I_c (at $H=0$ and $T=30$ mK) for five samples (filled squares). The samples with $R_n=70$ and 110 kΩ were the smaller junctions in two-junction arrays. Their areas were $\simeq 0.04$ and 0.05 μm^2, respectively. The samples with $R_n=15$, 35, and 140 kΩ were single junctions, with areas of approximately 0.1, 0.1, and 0.05 μm^2, respectively (see Fig. 4, inset). These estimates have an accuracy of roughly ± 0.01 μm^2. The A-B line is the Ambegaokar-Baratoff critical current prediction. The other two lines are our estimate (Ref. 2) $I_c = \pi e E_J^2/(8\hbar E_c)$, plotted for two reasonable capacitance values.

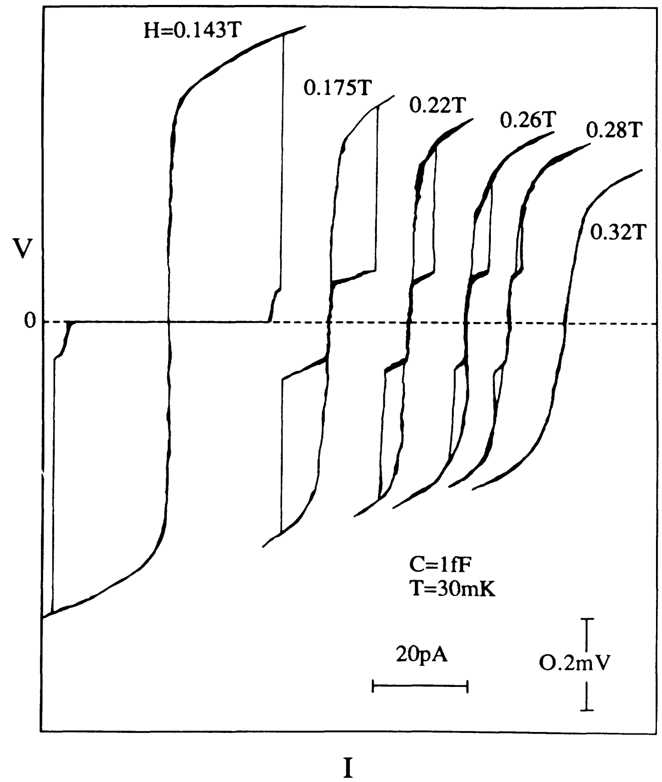

FIG. 3. Series of I-V curves taken as a function of magnetic field, for the sample with $R_n = 140$ kΩ.

preciable,[11,12] and the classical description breaks down.

We begin our discussion with E_c of order E_J. In this regime, the phase uncertainty is already comparable to the well separation in the Josephson potential $(-E_J \cos\phi)$, and quantum tunneling of ϕ is the dominant low-temperature phase-slip mechanism even at zero-bias current. We have previously reported[2] two new phenomena in such samples. First, the measured critical current follows a dramatic nonmonotonic temperature dependence[2] and is greatly reduced below the Ambegaokar-Baratoff[13] (A-B) value, even at the lowest accessible temperatures. Second, the I-V curve shows an anomalous resistance R_0 for $I < I_c$. We have developed a simple model[2] in which the quantum mechanical spread in the wave function $\psi(\phi)$ causes a reduced $I_c \propto E_J^2/E_c \propto C/R_n^2$, for $T \rightarrow 0$. The data shown in Fig. 2, recently obtained on a wider range of samples than was available in Ref. 2, confirm the R_n^{-2} dependence[14] predicted there and agree in absolute magnitude with our estimates. By modeling R_0 as the consequence of a series of phase tunneling events relaxed by damping, we have also recently obtained semiquantitative agreement with the measurements.[15]

In order to study the crossover behavior as the charging energy becomes more dominant, it is advantageous to be able to vary E_c/E_J continuously *in situ*. To do this, we have applied a magnetic field parallel to the substrate. The field reduces E_J both by phase modulation[16] and by reducing the superconducting energy gap. In our junctions, the magnetic field decreased I_c monotonically; the absence of a $\sin(x)/x$ dependence is probably due to a nonuniform barrier in the device.

Figure 3 shows a sequence of I-V curves taken as a function of applied magnetic field. While the low-H behavior is consistent with our earlier zero-field results, when H is increased above a threshold field H_t ($\simeq 0.16$ T in Fig. 3), a new regime is found. The response is *highly* resistive at small currents ($R \simeq R_L \sim 10^8$ Ω), steeply rising to a plateau at $V = V_b$. Thereafter V rises on a gentle ramp with slope $R \sim 10^6$ Ω, from which it exhibits a sharp voltage jump up to the gap voltage V_g at a current that we identify as I_c. At $H \sim H_t$ we estimate that E_c/E_J has reached ~ 50, using our $T=0$ estimate $I_c \propto E_J^2/E_c$ to infer the reduction of E_J from the observed reduction in I_c. I_c appears to vary continuously between the low-field and high-field values, suggesting the interpretation that it is still due to Josephson tunneling in the new regime. H_t depends on temperature, as shown in Fig. 4 (for a different sample). As temperature increases, E_J decreases and, accordingly, the magnetic field required to induce the new regime decreases. H_t also depends on the size of the sample, varying at low T from about 0.085 T in our $R_n = 15$ kΩ sample (Fig. 4), to about 0.16 T in our smaller-area 140-kΩ sample (Fig. 3).

Once the novel behavior sets in, the value of V_b does not appear to depend strongly on H or T. The value of

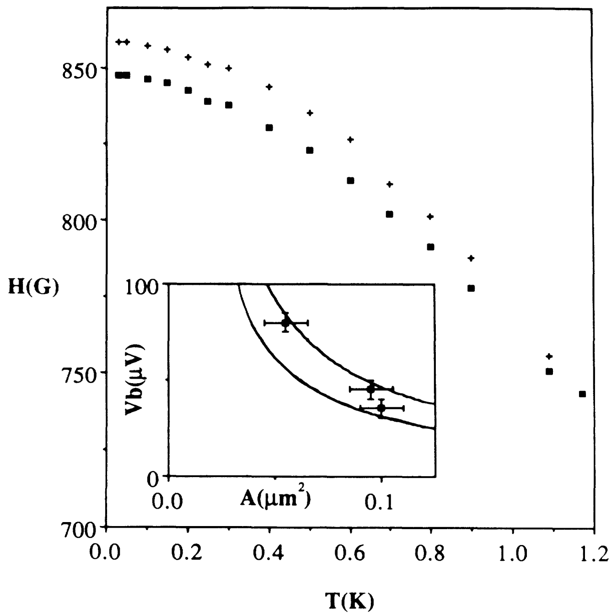

FIG. 4. State diagram indicating the region in which the knee at V_b was observed in a sample with $R_n = 15$ kΩ and $C \simeq 2$ fF. The filled squares correspond to H_t, and the crosses correspond to the field at which the plateau disappears. Inset: The measured blockade voltage V_b as a function of junction area. The three measured samples (from left to right) had $R_n = 140$, 35, and 15 kΩ. The curves correspond to $e/2C$, calculated with an oxide barrier thickness of 30 Å (top curve) and 20 Å (bottom curve), and $\epsilon_r = 6$.

V_b corresponds to the estimated $e/2C$ for each of the single-junction samples measured, as shown in the inset of Fig. 4. We estimate the capacitance from the junction area, obtained from SEM photographs, using a dielectric constant ($\epsilon_r \sim 6$) typical[17] of SnO_x barriers grown by glow discharge. The barrier thicknesses implied by the fit shown in the inset are reasonable for SnO_x and correlate with the oxidation times used for different samples. The values of C consistent with our interpretation of V_b are also in satisfactory agreement with those required in our interpretation of the $H=0$ measurements, both in the low-T quantum regime outlined above, and in the semiclassical regime at higher T.[2]

We propose the following simplified phenomenological picture: At voltages below $e/2C$, the tunneling of electrons (singly or as Cooper pairs) is energetically unfavorable and is inhibited, yielding a static situation; charge is trapped on the junction electrodes and the resistance is extremely high. At voltages greater than $e/2C$, on the other hand, single-electron tunneling is energetically favorable, and the differential resistance decreases, giving rise to a knee in the I-V curve at that voltage. In this dynamic regime, if the instantaneous voltage increases beyond e/C, it becomes energetically favorable for Cooper pairs to tunnel, and the voltage is driven back down. The observed dc voltage is thus restricted to a value below e/C, until the system's ability to transfer Cooper pairs is exceeded at $I = I_c$.

Several authors have investigated theoretically the behavior of a Josephson junction in the large E_c/E_J regime.[9,10,18–21] These treatments predict I-V curves which, because of the delocalization of the wave function $\psi(\phi)$, are highly resistive at small bias currents, as are the ones observed. Discrepancies exist, on the other hand, such as the absence in our I-V curves of the predicted negative-resistance region. We believe, however, that our simple interpretation may involve the same physics as more sophisticated models, such as the one by Guinea and Schön,[20] who derive an energy-band spectrum for a Josephson junction. To restate our previous explanation in the language of the band model, the highly resistive part of the I-V curve may then be due to the system being trapped at a fixed charge on the lowest band. This configuration is, however, only stable for $V < e/2C$. At higher (average) voltages, the system must spend time in higher bands and tends to relax to lower bands by tunneling, thus conducting charge and reducing the differential resistance of the device. This happens until the current is large enough for Zener processes to become so likely that the Josephson band gaps are ineffective at keeping the system in low bands. At this point ($I = I_c$) the voltage rises sharply to the energy gap.

Before a quantitative comparison with theory can be made, important issues must be resolved: First, in order to include the effect of damping, one must establish whether the damping resistance can be treated as linear (and, if so, equal to R_L, R_n, R_0, the real part of the effective impedance of the leads, or some combination of these), or whether the BCS quasiparticle response (nonlinear in frequency and ϕ) should be used. Furthermore, a realistic model for the actual current source should be used, that accounts for its intrinsic capacitance and inductance. Finally, when E_c is large, but E_J is still significant, the characteristic time scale of the junction dynamics is not well understood. When $E_c \ll E_J$, the Josephson plasma frequency (typically of order 10^9–10^{11} Hz) is the characteristic frequency of the system; in the opposite regime, when the charging energy is completely dominant, the characteristic frequency is thought[3] to be the inverse of the tunneling time, typically of order 10^{15} Hz. If the time scale is sufficiently fast, the stray capacitance affecting the junction will be small.

Some information on these damping and stray capacitance issues can be gained by comparison of results on our samples in single-, double-, and eleven-junction configurations. Unfortunately, the magnetic field measurements could only be performed successfully in single-junction samples; in more complicated devices, unless all junctions are identical, different junctions will have different magnetic field responses, greatly complicating the interpretation of the data. However, the

$H=0$ measurements on different sample configurations are all consistent (as indicated in Fig. 2, for example); thus the observed low-temperature behavior of the smallest junction in a given sample appears unaffected by the presence of companion junctions in the leads, although their series Josephson inductance should greatly affect the lead impedance. Moreover, as noted above, we have fitted our data on I_c and R_0 in different regimes, using quantum tunneling models at low temperature, and classical models near T_c.[2,15] While the models used are quite simplified, the dynamics of our devices appear in all cases to be determined by their *intrinsic* capacitance and resistance, although the possible contribution of a *small* amount of parasitic capacitance (<1 fF) cannot be excluded. This independence of lead configuration and stray capacitance is in contrast with the observations of Devoret *et al.*[11] whose macroscopic quantum-tunneling measurements were affected by the impedance of their leads. A possible explanation for this difference is that as E_c becomes important (the value of E_c for the junctions used in Ref. 11 was $\simeq 10000$ times smaller than for ours), the tunneling process may become more local and microscopic in nature (and the time scale fast), because of the dominance of single-electron fluctuations.

As H is increased further (to $H=0.32$ T in Fig. 3), the Josephson-type I_c is suppressed, and the plateau in the I-V curve disappears. With further increase in the field, the junction eventually becomes normal, as the superconducting energy gap $\Delta \rightarrow 0$ (at $H \sim 1$ T for the sample in Fig. 3 with $R_n = 140$ kΩ). The I-V curve at this field is still nonlinear, with an offset voltage corresponding to the Coulomb-blockade voltage $e/2C$. As the field is increased still further, however, the nonlinearity in the I-V curve washes out at about 2 T. We suggest that at this point $\tau_{\mathrm{s.o.}}(\mu_B H)^2/\hbar \sim E_c$ (where $\tau_{\mathrm{s.o.}}$ is the spin-orbit scattering time of the electrons, and μ_B the Bohr magneton), and the Coulomb blockade is rendered ineffective by level broadening by spin-alignment energies in the presence of spin-orbit scattering processes.[15]

In conclusion, we have described the experimental behavior of small tunnel junctions as the ratio of charging energy to Josephson energy is increased. At moderate Josephson energies (such that $E_J \sim E_c$), the behavior of the Josephson device is modified, the I-V curve being resistive for all currents, and the critical current being greatly reduced even as $T \rightarrow 0$. The significant quantum uncertainty in ϕ allows for tunneling events between wells of the Josephson potential. At very small Josephson energies, such that $E_J \ll E_c$, the whole character of the I-V curve is changed. Although the electrodes are still superconducting, tunneling appears almost completely inhibited until the voltage difference between the electrodes is sufficient to make the tunneling of one electron energetically allowed. In this regime the charge can be treated semiclassically; hence, the phase wave function is highly delocalized and no longer effectively pinned by the Josephson potential.

The authors gratefully acknowledge the generous help of Walter F. Smith in carrying out the dilution refrigerator measurements. This research was supported in part by National Science Foundation Grant No. DMR-84-04489, U.S. Office of Naval Research Contract No. N00014-83-K-0383, and by Joint Services Electronics Program Contract No. N00014-84-0465.

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[14]It may be significant in relating our low-temperature observations to those of Orr *et al.* on granular films (see Ref. 8) that an extrapolation of our observed $I_c \propto R_n^{-2}$ dependence intersects the $I_c \propto R_n^{-1}$ dependence of A-B at $R_n \sim 6$ k$\Omega \sim R_Q = h/4e^2$. Moreover, for $T \ll T_c$, we find a significant R_0 only in samples with $R_n > R_Q$.

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[16]We assume that E_J is reduced by a magnetic field just as I_{c0} is known to be. This is confirmed by experiments (see Ref. 11), in which the plasma frequency $\omega_p = (1/\hbar)(8E_J E_c)^{1/2}$ reduced by a magnetic field.

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