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# Superconducting energy gap in Coulomb staircase tunneling structures

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A sharp Coulomb staircase $I$-$V$ characteristic has been observed in tunneling structures formed between a tungsten scanning tunneling microscope (STM) tip and a superconducting lead film. The actual geometry is that of a single isolated particle acting as a common electrode in a series configuration of two low-capacitance tunneling junctions between the bulk of the lead film, the particle, and the STM tip. The voltage width of the step that crossed zero bias was larger than that of other steps in the staircase. This is due to the superconductivity of lead, and is consistent with theory.

Nearly twenty years ago, electrostatic charging was observed to strongly affect $I$-$V$ characteristics[1] and other electrical properties of very small (mesoscopic) tunneling structures.[2] Several phenomena may occur when the macroscopic charging energy is larger than thermal energies. In a current-biased junction, a voltage threshold for conduction resulting from a Coulomb blockade has been predicted[3] and experimentally observed.[4-6] In a series combination of a capacitor and a tunnel junction, nonlinear capacitance effects are found.[2,7] In a series combination of two tunnel junctions the so-called Coulomb staircase pattern in the $I$-$V$ tunneling characteristic is possible.[8,9] The latter has been observed in geometries in which a junction pair is formed by a single particle juxtaposed between two electrodes[10] and in random parallel combinations of such junction pairs.[11,12] Superconductivity should modify the staircase[13] as it does in the case of the Coulomb blockade voltage threshold.[4] Although one observation of the Coulomb staircase other than in high-$T_c$ superconductors[10] did involve superconducting electrodes,[12] no experimental data has yet been available that directly demonstrates the theoretical predictions. Here we report the first observation of a modification of the Coulomb staircase of a mesoscopic tunneling structure directly attributable to the superconductivity of the materials involved.

An ideal structure for observing the Coulomb staircase is one in which the current is limited to a single channel and is carried by single electron tunneling (SET). In this instance the effect would not be smeared as is the case in the random systems[11,12] consisting of more than one conducting channel, with different junction parameters. A structure in which SET is realized may be useful for observing related effects that have been predicted such as SET voltage oscillations in current biased junctions[14] or Bloch oscillations[15] in superconducting junctions.

A 2000-Å Pb film was vapor deposited through a $10^{-5}$-Torr $O_2$ atmosphere and onto a liquid-nitrogen-cooled sapphire substrate.[16] The resultant films consisted of Pb grains of various sizes separated by natural oxide barriers. Magnetic-susceptibility measurements using a superconducting susceptometer showed the onset of a Meissner effect at 7.0 K. Tunneling structures were formed between the Pb film and the tip of a low-temperature scanning tunneling microscope (STM). For the purpose of further discussion we consider a two-junction model of this structure. Junction 1 refers to the junction between the STM tip and the Pb particle on the film nearest the tip. For simplicity, we disregard the complexities of the rest of the Pb film and simply refer to it as the "bulk of the film." Junction 2 refers to the junction between the aforementioned Pb particle and the bulk of the film.

The STM (a tubular piezoelectric scanner design) and Pb film were cooled to 4.2 K by immersion in liquid helium. Tunneling characteristics were obtained by turning off the STM feedback for time intervals of 85 ms during which the bias voltage was ramped to the opposite polarity and back to its original value. In order for the STM feedback circuit to be effective in controlling the height of the tip above the film, the tip to particle junction (junction 1) must be the current limiting junction, i.e., $R_1 \gtrsim R_2$, where $R_1$ and $R_2$ are the resistances of junctions 1 and 2, respectively. For all of the data presented here, we insured this criterion by being certain that the tunnel current was both equal to the feedback set-point current and sensitive to changes in the set-point value. The main advantage of using a STM to form the structure is that SET may be achieved by selecting a single Pb particle as an intermediate site in the tunneling from the tip to the film. Additional advantages are that junction 1 can be made very small resulting in $C_1$ being small, two of the parameters ($R_1$ and $C_1$) can be varied by changing the STM tip height above the film, and the tip can be moved to various spots on the film to vary $R_2$ and $C_2$. The STM tip was sharpened tungsten wire which was allowed to touch the film, resulting in a Pb coating and effectively producing a Pb tip. The evidence for this was the fact that the $I$-$V$ and $dI/dV$-$V$ characteristics taken at a spot on the film that did not exhibit a staircase showed well-defined BCS gap features consistent with a superconductor-insulator-superconductor junction, rather than a superconductor-insulator-normal configuration.

At several locations on the film, the tunneling characteristic exhibited a staircase pattern as shown in Fig. 1. The plot of $dI/dV$ vs $V$ (also shown in Fig. 1) has a peak for each stepwise increment in the current. The observation of a sharp staircase is evidence for the claim that elec-

 

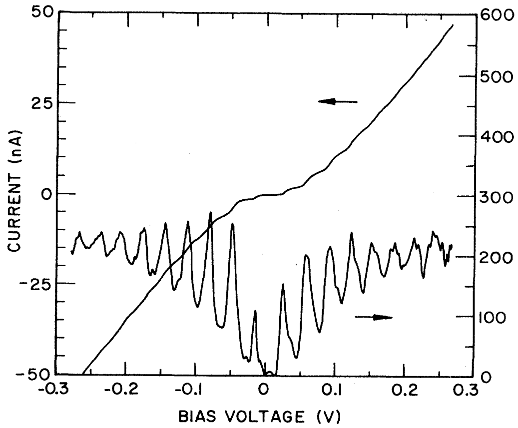


FIG. 1. Plots of $I$ vs $V$ and $dI/dV$ exhibiting a clear Coulomb staircase. These were obtained from a tunneling structure formed between a granular Pb film and a STM tip.

tron tunneling through a single isolated Pb particle (SET) is occurring. The lack of smearing is evident from the sharpness of the steps in the staircase and the very large oscillations in $dI/dV$. The sharpness of the pattern suggests that $R_1 \gg R_2$, $C_1 \gg C_2$, and $e2/C_1 \gg kT$ for this structure.[9] The circles in Fig. 2 show how the width of a step depended on the bias voltage at the center of the step for the $I$-$V$ shown in Fig. 1. The voltage at the step center was taken to be the average voltage of two adjacent peaks in $dI/dV$. For the most part, the current increments and corresponding $dI/dV$ peaks were equally spaced resulting in equal step widths. The width of the step that crosses zero-bias voltage was wider than the other steps by 10 mV. In contrast, for structures consisting entirely of normal metals, the width of *each* step of the Coulomb staircase has been predicted[9] and observed[10,11] to be $e/C_1$

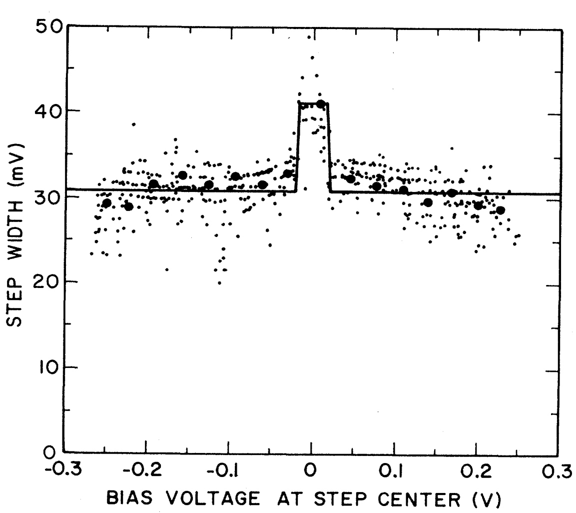


FIG. 2. The circles show the step width versus the bias voltage at the center of the step for the Coulomb staircase shown in Fig. 1. The dots show similar data obtained from 39 additional staircases. The steps crossing zero bias were 10 mV wider than the others.

(assuming $C_1 > C_2$). Forty $I$-$V$ curves (one of which is shown in Fig. 1) were taken at time intervals spaced thirty seconds apart without changing the tip position. The dots in Fig. 2 show the variation of the width of a step depended on the bias voltage at the center of the step for the steps in these $I$-$V$ curves. Although there was scatter, the steps crossing zero bias were generally 10-mV wider than the other steps. The widths $\delta V_{\text{step}}$ were found to depend on the voltage at the center of the step $V_{\text{step center}}$ as

$$\delta V_{\text{step}} = \begin{cases} e/C_1 + 8\Delta, & |V_{\text{step center}}| < e/2C_1 , \\ e/C_1, & |V_{\text{step center}}| > e/2C_1 + 4\Delta . \end{cases} \quad (1)$$

For this structure $e/C_1$ was 31 mV and $8\Delta$ was 10 mV. The value of the parameter $\Delta$ in Eq. (1) is the superconducting gap because for a series combination of two junctions with all electrodes superconducting, the likely arrangement realized in this instance, theory predicts that the width of the step crossing zero bias is wider than the other steps in the staircase by eight times the superconducting gap.[13] The fact that we have performed the same experiments on 300-Å thick nonsuperconducting Au films and found Coulomb staircases of similar quality to that of Fig. 1 without an enhanced width of the voltage step crossing zero bias supports this assertion. The magnitude of the Pb gap, $1.25 \pm 0.1$ mV, inferred from this interpretation of the data was consistent with the known Pb gap and provides a demonstration of the predicted modification of the Coulomb staircase by the superconductivity in Pb at 4.2 K.

The step edges, the voltages at which current increments occur, as shown in Fig. 1, are given by

$$V_{\text{step}} = Q_0/C_1 + ne/C_1 + 4\Delta\,\text{sgn}(Q_0/C_1 + ne/C_1) , \quad (2)$$

where $\text{sgn}(x) = x/|x|$ and $n$ is an integer. $Q_0$ may be interpreted as a charge on the capacitor introduced by polarization of the dielectric. The latter two terms of this equation contain the same information as Eq. (1). The first term determines the voltage for the first step. The $ne/C_1$ term in this equation allows us to limit $Q_0$ to the range $-e/2 < Q_0 \leq e/2$ without loss of generality. The significance of $Q_0$ has been investigated theoretically[8] and convincingly demonstrated in microfabricated structures that did not have a Coulomb staircase.[4] Frequent observations of $I$-$V$ traces that followed Eq. (1) showed extremely well that traces could be acquired in which $Q_0$ did not change during the acquisition of the trace. The value of $Q_0$ varied between traces even when the traces were taken only seconds apart and without changing any parameters within our control. This is illustrated in Fig. 3. In addition, $Q_0$ sometimes changed once or twice during a trace. These changes were marked by an abrupt increase or decrease in current that was smaller than the current increment of an ordinary step. This resulted in steps whose width in voltage was anomalously short or long. These occasional anomalies in step width cannot be attributed to errors in the location of the conductance peaks, because if this were the case, an anomalously short step would occur adjacent to an anomalously long one which was not found. Two effects of the abrupt changes in $Q_0$ are apparent in Fig. 2. The centers of the steps are uni-

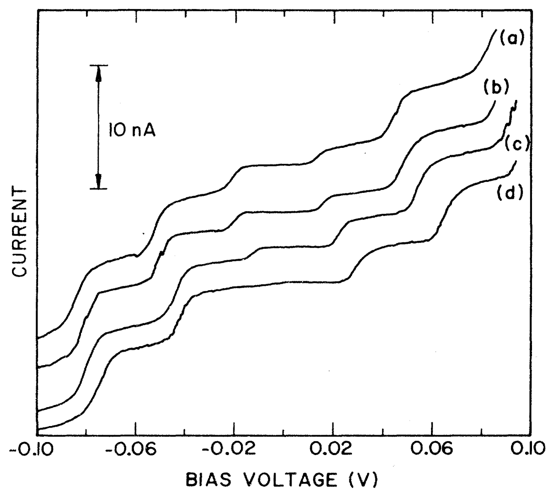


FIG. 3. Traces (a)–(d) show four different Coulomb staircase $I$-$V$ characteristics with the STM tip in the same position. The traces are displaced vertically for clarity. Although the step widths are approximately the same for each trace, the steps occur at different bias voltages, that is, they have different values of $Q_0$. [See Eq. (2).] In (d) it is probable that a step edge occurs close to zero bias and is not resolved in this trace because the step height is too small.

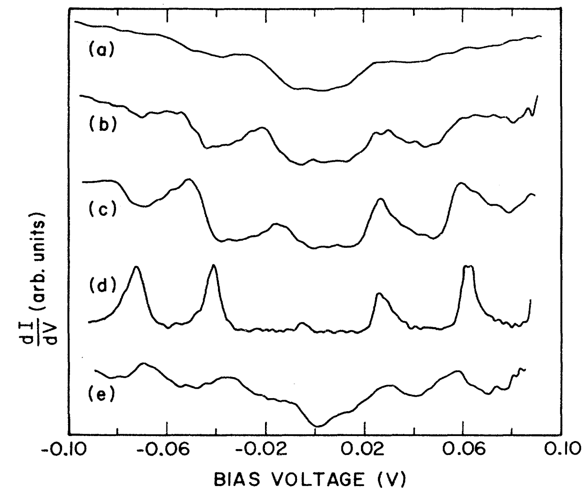


FIG. 4. (a)–(e) Coulomb staircase $dI/dV$ vs $V$ characteristics corresponding to increasing proximity of the STM tip to the Pb film. The curves are separately normalized and offset for clarity. The values of the junction resistance in $M\Omega$ at 75 mV (which serves as a measure of tip height) for (a)–(e) are 66, 26, 17, 9.6, and 7.1, respectively. The peak separation (step width) decreased with increasing proximity.

formly distributed along the voltage axis, and the scatter in step widths is larger than the measurement error. The previously reported "bistability" in the Coulomb staircase[10] can be described by Eq. (2) as having $Q_0$ change between the values of 0 and $e/2$. Changes in $Q_0$ have been attributed to the random filling of traps with electrons resulting in changes in the polarization of the dielectric.[10] Our results are consistent with this explanation.

Figure 4 shows that the staircase pattern was sensitive to $d$, the STM tip height above the Pb particle. Increasing the tip height caused the step width to increase due to the increase in $e/C_1$. When the tip was far from the film (a), the staircase was smeared due to the breakdown of the requirement $C_1 \gg C_2$. When the tip was very near the film (e), the staircase was smeared due to breakdown of the requirement $R_1 \gg R_2$ (see Ref. 11). The variation of the step width and the staircase sharpness with STM tip height is strong evidence that the discussed charging effects in the two junction model causes the observed $I$-$V$ characteristics.

The current increments $\delta I_{\text{step}}$ would be equal and given by $\delta I_{\text{step}} = \delta V_{\text{step}}/R_1 = e/C_1R_1$ if junction 1 had a linear current-biased $I$-$V$; however, this is modified because the Coulomb blockade gives junction 1 a nonlinear current biased $I$-$V$ characteristic. If the steps are sufficiently flat, the result can be stated graphically as the requirement that the centers of each step, except that crossing zero bias, lie along the Coulomb blockade $I$-$V$ characteristic for a current biased junction equivalent to junction 1. Our results were consistent with this. One consequence of this is that a step near zero bias may have a step height that is too small to see. Because of this we inferred that a step near zero bias was present if current increments occurred near $e/C_1 + 4\Delta$ and $-e/C_1 - 4\Delta$. Figure 3(d) shows such a case.

In conclusion, we have observed a sharp Coulomb staircase in tunneling structures involving a STM tip and a granular Pb film. The results are consistent with the structure modeled as two superconducting tunneling junctions in series. The extra width of the step crossing zero bias can be attributed to the superconducting gap in Pb. This effect is similar to the augmentation of the Coulomb blockade voltage threshold by the superconducting gap as reported by Fulton and Dolan[4] in lithographically produced structures.

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