POLYTECHNIC UNIVERSITY

DEPARTMENT OF PHYSICS

CONTROL OF THE RESIDUAL SUB-ELECTRONIC CHARGE ON A MESOSCOPIC CONDUCTOR BY MEANS OF A SCANNING TUNNELING MICROSCOPE TIP

Final Technical Report, ONR Research Grant N00014-90-J-1845

Grant Title: " Experimental and Theoretical Studies of Coulomb Blockade Phenomena in Mesoscopic Normal State Systems" , E. L. Wolf PI, Department of Physics, Polytechnic University, Six Metrotech Center, Brooklyn, NY 11201

Prepared March 7, 1994 by E. L. Wolf E L Wolf

I. Introduction

The topic of research has been the Coulomb Blockade (CB) as observed in cryogenic Scanning Tunneling Microscopy using a voltage biased tip. The basic Coulomb Blockade effect is the freezing out of stepwise hopping conduction of electrons through sites where the classical Coulomb energy, $e^2/2C$, for site occupation, is unavailable both thermally and from the bias source. In our experiments the blocked sites reside on small metallic particles deposited on, but slightly decoupled from, the underlying metallic or superconducting substrate.

A single tunnel junction with very small capacitance C under current bias at low temperature has been shown in the theory of Averin and Likharev [1] to exhibit a smooth Coulomb Blockade (CB) I(V). Here I is quadratic in V, the time average junction voltage, below the classical Coulomb charging threshold for one electron, $V_c = e/2C$, with a linear I(V) at higher voltages. In the blockade regime, $V << V_c$, current limited tunneling of single electrons across the junction is nearly periodic, with period e/I. The instantaneous junction voltage varies linearly between +/- e/2C interrupted at time e/I by an abrupt electron tunneling event. In this single junction case, only strict current bias will allow observation.

Although reports of STM-based experimental observations have been given, these may inadvertently have involved a small particle, making the configuration in fact a double junction, series connected. Microlithographically formed structures are more credible in achieving the conditions for observation of the single junction Coulomb Blockade case.

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March 14, 1994

Dr. Larry R. Cooper
Code 1114SS
Office of Naval Research
800 North Quincy Street
Arlington, VA 2217-5000

RE: Final Technical Report for Grant N00014-90-1845

Dear Dr. Cooper:

I have attached three copies of the Final Technical Report for the above Grant entitled: "Experimental and Theoretical Studies of Coulomb Blockade Phenomena in Mesoscopic Normal State Systems". This Grant did not involve acquisition of Government Furnished Equipment. No disclosures, inventions or patents resulted. Also enclosed are reprints of the principal publications resulting from the Grant.

Please let me know if further information should be required.

Sincerely yours,

E L Wolf

E. L. Wolf
Professor and Head

Copies: (1) Administrative Grants Officer, Office of Naval Research
Resident Representative N62927
Administrative Contracting Officer
33 Third Avenue--Lower Level
New York NY 10003-9998

(1) Director, Naval Research Laboratory
Attn: Code 2627
Washington DC 22314

(4) Defense Technical Information Center
Building 5, Cameron Station
Alexandria, Virginia 22314

(1) Joseph Scelso, Polytechnic

However, the larger size of the available lines makes the temperature scale necessarily much lower than the 1K range adequate for STM based work.

The double junction case, studied with an STM, is the subject of the supported research. The series connection of two small capacitance tunnel junctions under voltage bias (double junction case) leads to the "Coulomb Staircase" I(V). This case was first analyzed by Giaever and Zeller [2], who studied I(V) of a tunneling capacitor whose dielectric was embedded with small metal particles. The Giaever-Zeller data, which did not actually reveal the staircase, was fit as the sum of parallel currents, the I(V) through each particle exhibiting steps of spacing e/C. Each particle corresponds to series connection of two small tunnel capacitances; the nth step gives average occupation of n surplus electrons on the inner electrodes (small particle).

This case was also studied experimentally in conventional tunnel junctions by Barner and Ruggiero [3], who resolved the staircase even in the parallel array; by van Bentum et al [4], and by Wilkins et al [5] with an STM tip to tunnel into individual Sn particles deposited upon a slightly oxidized Al surface. Prominent steps were observed [7] whose spacing was interpreted as e/C where C is the larger of the two capacitances of drop to tip and sample, and corresponds also to the junction of larger R. Shunting lead capacitances do not cause a problem in this case, since a genuinely small capacitance on order 10^{-18} F exists between the particle and its surroundings.

II. Experiments

Our STM based studies of the CB involved a mesoscopic 100Å evaporated particle, usually gold, evaporated upon, but slightly decoupled from, a conducting substrate. Electrons hop (tunnel) from the tip to the particle and later hop to the substrate. The two junctions formed have exceedingly small capacitances, on the order of 10^{-18} F, by the small size of the particle. This configuration leads to the "Coulomb Staircase" form of I(V) characteristic, clearly observed directly in plots of the I-V curves. The basic voltage spacing of the steps is e/C, where C is related to the tip-particle, and particle-substrate capacitances. In our work the field of particles was observed using the STM in the topographic mode, and detailed measurements of the I(V) curves **with the STM tip at varying heights** above the particle were acquired.

The STM is a single piezo tube scanner [6] with tip and sample in liquid helium. The design has been modified to improve thermal compensation and the unit is mounted in an acoustic chamber on an air suspension.

To produce the small particles, extremely thin films of 99.998% gold were evaporated from a Ta boat at $1X10^{-6}$ torr onto freshly cleaved samples of the high temperature Bi 2212 superconductor, or onto gold films pre- evaporated onto mica in thermal contact with a copper heat sink at room temperature. The evaporation rate was 10-15 Å per second and the thickness of the Au film was typically 7000 Å (for a base film) or 10 Å (to produce particles), as measured by a quartz microbalance. STM topographs show grains of several hundred Å size on which atomic steps and terraces are occasionally observed. The Au films had been exposed to air for periods ranging from hours to days, while the Bi 2212 samples were freshly prepared by cleaving (peeling).

W tips were made by electrochemically etching 0.25mm diameter W wires at 10 V DC in 10% KOH solution using a 1/8 inch diameter loop of stainless steel wire as counterelectrode. The W tips were rinsed in water and acetone after etching, immediately prior to the experiment.

III. Results

In an initial study of Au films without intentionally deposited small particles, most of the I-V data were linear. Therefore we searched for the infrequent Coulomb Blockade I(V) curves in raster scans of 10^4 points, in a 2000 A square on the Au surface. At each raster point, I-V curves were acquired at 5 different current levels (tip-sample spacings). At an initial tip bias V_0 (typically 300 mV), with current fixed at the desired initial level by the feedback, the servo is disabled, and V swept linearly to $-V_0$ and return, acquiring 100 $I(V_i)$. In some cases the process was repeated at the same raster location, after restabilization of the tip height to $I(V_0)$. One run takes 3-4 hours. The data file created in a typical raster scan, about 20 megabytes, includes a 100X100 topography matrix, with 5 I-V curves (200 points each) for each raster point. In some cases nonlinear I(V)'s were studied under manual control at more than 5 tip-sample spacings.

In other cases, especially using the Bi 2212 superconductor as substrate, the tip was located at a point on the sample where the staircase I(V) was observed, and then a computer-controlled measurement process was used wherein 50 different tip heights were scheduled, and at each tip height (corresponding to a distinct tip-particle capacitance) more then 5 I(V) curves were acquired for later analysis.

The most interesting new observation from our work was that the tip-particle spacing, expected vary the related capacitance, **also caused the "Coulomb Staircase" pattern to shift along the voltage axis.** The interpretation of this is that the non-integer residual charge on the particle is being controlled by the tip spacing. It was found that a reasonable difference between the work function of the tip and of the small particle could account for the observed shifts. An algorithm was developed to model the data obtained, and from this it was also possible to determine the superconducting energy gap of the underlying Bi 2212 superconductor. This work was presented orally at the International STM Conference and was published [P1] in the Proceedings of that Conference. A more complete version of the work was published [P2] in the IEEE Transactions on Magnetics. The work was also presented at the March 1991 APS Meeting in Cincinnati OH [P3].

SUMMARY

In summary, capacitive control of the residual non-integer electric charge on a mesoscopic conductor has been demonstrated by varying the spacing of an STM tip, presumed to have a different work function than the mesoscopic conductor. The demonstrated resolution of this control is on the order of e/100, or 1.6 x 10^{-21} Coulomb.

REFERENCES

[1] D. V. Averin and K. K. Likharev, J. Low Temp. Phys. 62, 345 (1986).
[2] H. R. Zeller and I. Giaever, Phys. Rev. 181 789 (1969).
[3] J. B. Barner and S. T. Ruggiero, Phys. Rev. Lett. 59, 807 (1987).
[4] P. J. M. van Bentum, H. van Kempen, L. E. C. van de Leemput, and P. A. A. Teunissen, Phys. Rev. Lett. 60, 369 (1988); P. J. M. van Bentum, R. T. M. Smokers, and H. van Kempen, Phys. Rev. Lett. 60, 2543 (1988).
[5] R. Wilkins, E. Ben-Jacob, and R. C. Jaklevic, Phys. Rev. Lett. 63, 801 (1989).
[6] A. P. Fein, J. R. Kirtley, and R. M. Feenstra, Rev. Scient. Instrum. 58,1806 (1987).

PUBLICATIONS

P1. Z. Y. Rong, A. Chang, L. F. Cohen, and E. L. Wolf, "STM control of the non-integer charge on a metal particle inferred from Coulomb staircase", Ultramicroscopy 42-44, 333 (1992)

P2. Z. Y. Rong, A. Chang, L. F. Cohen, and E. L. Wolf, "STM Control of Noninteger Residual Charge Q_0 on the Central Electrode of a Double-Tunnel Junction", IEEE Transactions on Magnetics 28 67 (1992).

P3. Zhao Yan Rong, Alejandro Chang, Lesley F. Cohen, and E. L. Wolf, "STM Control of the Non-integer Charge Q_0 on the Central Electrode of a Double Tunnel Junction", Bull. Am. Phys. Soc. 36 873 (1991).

EQUIPMENT

No Government Furnished Equipment was acquired under this grant.

PERSONNEL

This grant provided partial salary support for the following persons:

Prof. E. L. Wolf, One summer month plus small fraction of academic year salary

Prof. S. C. Meepagala, small fraction of academic year salary

Dr. Lesley Cohen, Postdoctoral, approximately 50% support over one year

Hong-jie Tao, Visiting Research Scientist, four months support

Zhao Yan Rong Graduate Student Research Assistant, four months support

Farun Lu Graduate Student Research Assistant, 1.5 months support

Ms. Roza Kotlyar, Undergraduate Research Assistant, hourly during summer

These have turned out to be very good research personnel, who have benefitted from the grant. Prof. Meepagala is being recommended for promotion to Associate Professor in our Department. Dr. Cohen is now with the Centre for High Temperature Superconductivity at the P. M. S. Blackett Laboratory, Imperial College, London, and has won a prestigious five year Royal Society (London) University Fellowship. Tao has returned to his permanent position at the Institute for Physics, Academy of Sciences in Beijing. Dr. Rong obtained his Ph. D. with partial support of the present grant, and is presently at State University of New York at Stony Brook, as a Postdoctoral Associate of Prof. Konstantin Likharev, a very prominent physicist. Dr. Lu is presently a Postdoctoral in my group at Polytechnic. Ms. Kotlyar got her BS in Physics and is now a graduate student in Physics at the University of Maryland.

APPENDIX: The Publications 1,2 listed above are appended.