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Coulomb blockade and an electron in a mesoscopic box

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I. SCOPE

These problems illustrate some properties of metallic grains so small that it is easy to observe the change in their electric potential when a single electron is added to or subtracted from a grain. The remarkable properties of such small systems may make possible more precise measurements of charge and current and may revolutionize information processing technology.

Solutions of the first few problems use familiar relations between energy, charge, voltage, and capacitance. They are problems suitable for introductory electricity. As examples of mesoscopic systems they may also be of interest for teaching modern physics. The problems connecting observed features to the energy levels of the electrons in the grain use the concepts of Fermi energy and density of states of electrons in a box and are nice applications of basic quantum mechanics.

II. INTRODUCTION

Using techniques and technology developed in the last 10 years, often referred to as "nanofabrication," we can now make solid structures of atoms that exhibit properties midway between the purely quantum behavior of a single atom interacting with an electron or two and the classical behavior of bulk matter. Such systems are called "mesoscopic" to suggest that they are intermediate between microscopic and macroscopic systems. Mesoscopic systems can show strong effects when just a single electron is added or subtracted. This sensitivity to individual electrons has led people to talk of "single electronics"[1] when the Coulomb energy controls the electron number with single-electron precision and of "artificial atoms"[2] when the discreteness of the quantum energy levels of the electrons is important. It is the prospect of developing such systems to the point where a single electron can carry one bit of information that has inspired talk of a revolution in computer technology.

We have used electron-beam lithography and reactive-ion etching to form a circular hole about 10 nm in diameter in a Si_3N_4 insulating membrane. As shown in Fig. 1(a), this tiny hole defines the area of a small tunnel junction which connects the flat side of a roughly hemispherical aluminum grain to an aluminum lead. The curved surface of the hemisphere is connected by a second tunnel junction to the other aluminum lead. The two aluminum leads connect the device to a voltage supply as shown in Fig. 1(b). The tunnel junctions have approximately 1-nm-thick oxide layers between the grain and the electrode and are modeled as leaky capacitors. The tunnel resistances act as ordinary linear resistances only at voltages well above the Coulomb blockade voltage discussed below. Below this voltage they are insulators and the equivalent circuit is essentially a capacitive voltage divider.

In an experiment the measured quantity is the current I through the device as a function of the voltage difference V

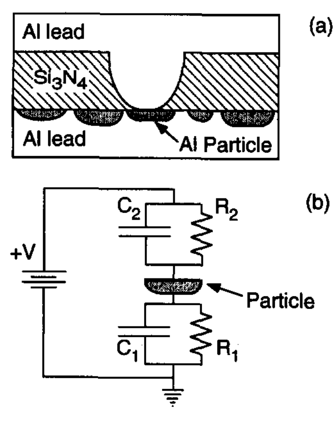

Fig. 1. (a) Sketch of the experimental configuration. The aluminum leads are thin films. The upper lead is deposited after the hole in the substrate has been fabricated. The lower lead is deposited after the granular film has been deposited and the surface of the grains oxidized. (b) Electrical equivalent circuit of the device. C_1 and C_2 are the capacitances between the particle and the leads, and R_1 and R_2 are the tunneling resistances through the oxide layer separating the particle from the leads.

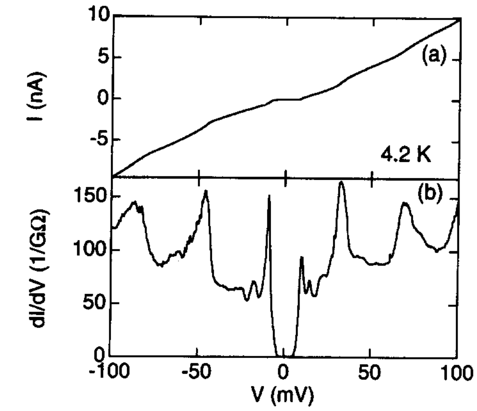

Fig. 2. (a) $I(V)$ curve of the device at 4.2 K showing the Coulomb blockade region with $I=0$ near $V=0$ and the inclined steps of the Coulomb staircase at higher voltages. (b) Plot of dI/dV, which makes these features more visible.

applied between the two electrodes. Measured $I(V)$ curves such as the one in Fig. 2 exhibit interesting effects. The flatness around $V=0$ is the result of what is called the "Coulomb blockade" of the current; the succession of steplike rises in the curve is called the "Coulomb staircase." Furthermore, with small enough grains and low enough temperatures, such data (Fig. 3) also exhibit the discrete energy states of electrons in the grain.[3] These are in essence the states of a particle in a box.

From analysis of such $I(V)$ measurements, we can determine the capacitance and resistance of a grain and its junctions. For the particular grain giving the data in Figs. 2 and 3, we found the capacitances of the two junctions 1 and 2 to be respectively $C_1=4.9\pm0.5$ aF, $C_2=8\pm1$ aF (where "aF" stands for "attofarad"$=10^{-18}$ F) and their tunnel resistances to be $R_1=8\pm1$ MΩ, $R_2=1\pm0.3$ MΩ. This grain will be used as a concrete example in some of the problems below.

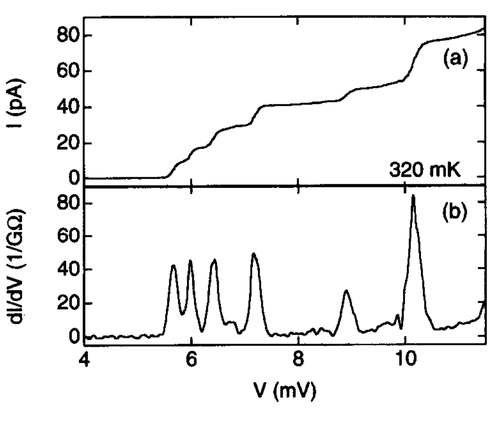

Fig. 3. (a) $I(V)$ curve of the device at 320 mK showing onset of current steps at voltages above the Coulomb blockade where individual quantum states in the grain become accessible to tunneling. (b) Plot of dI/dV, in which each onset step appears as a peak.

III. MAGNITUDES OF THE PHENOMENA

You can get a sense of the effect of a single electron on a mesoscopic system by estimating the capacitance of the system and calculating the change in its voltage when a single electron is added.

A. Magnitudes—problems

(a) Estimate the self-capacitance of an aluminum grain $\sim$10 nm in diameter if it is isolated in free space.

(b) Estimate the capacitance to ground of the same grain if it is enclosed by grounded metal films that are separated from the grain by an insulating oxide layer 1 nm thick with a dielectric constant of $\kappa=8$. The insulating layer is sufficiently thin that the arrangement can be modeled as a parallel-plate capacitor. Compare the capacitance with the result of (a) to get a sense of how much the electrodes and tunnel junctions affect the overall capacitance of the system.

(c) By how much would the voltage on such a grain change if a single electron were added to it in each of the above two cases?

(d) It is interesting to know the relative change in the number of free (conduction) electrons in the grain when you add a single electron. Use the fact that aluminum has three free electrons per atom to estimate the number of free electrons in such a grain.

B. Magnitudes—answers

(a) The potential V of an isolated conducting sphere of radius R carrying a charge Q is $V=Q/4\pi\epsilon_0 R$ where ϵ_0 is the permittivity of free space. Because $Q=CV$, $C=4\pi\epsilon_0 R$. Therefore,

$$C=\frac{5\times10^{-9}}{9\times10^{9}}=0.55\times10^{-18}\ \text{F},$$

that is, approximately 1 aF.

(b) The capacitance of a parallel-plate capacitor is $C=\kappa\epsilon_0 A/d$, where A is the area of the plates, d is their separation, and κ is the dielectric constant of the medium between the plates. Inserting $A=4\pi R^2$, you find that the capacitance exceeds the self-capacitance found in (a) by a factor of $(\kappa R)/d=8\times5/1=40$. Therefore, even if the electrodes cover only a fraction of the surface of the grain, their capacitance will dominate.

(c) The change in voltage caused by adding an electron, $Q=-e$, is

$$|\Delta V|=\frac{1.6\times10^{-19}}{0.55\times10^{-18}}=290\ \text{mV}$$

for the isolated grain,' or $290/40\approx7$ mV for the grain completely enclosed by electrodes. These are large, easily observed voltages.

(d) The volume of the grain is about $(10\ \text{nm})^3=10^{-24}$ m^3. The gram atomic weight of aluminum is 27 and its density is 2.7 g cm^{-3}. This means that a mole of Al occupies $27/2.7=10$ cm^3, and there are $6\times10^{23}/10$ Al atoms in a cubic centimeter or $3\times6\times10^{22}$ electrons in a cubic centimeter. Consequently, the grain contains $10^{-18}\times1.8\times10^{23}$ $=1.8\times10^5$ free electrons.

This result is characteristic of a mesoscopic system. The number of its components is large, but still small enough to exhibit strong effects from a single elementary particle. In this case, the addition of 1 electron to a collection of 10^5 produces a measurable change in voltage.

IV. DETERMINING GRAIN SIZE FROM CAPACITANCE

In an experiment the size of the grain is deduced from the capacitance rather than the other way around. For example, with a simple model of the shape of the grain it is easy to estimate the size of an aluminum grain that has the values $C_1 \approx 5$ aF and $C_2 \approx 8$ aF mentioned above in Sec. II. [These values were determined by fitting features of the Coulomb staircase in the $I(V)$ curve as will be discussed in Sec. V.]

A. Grain size—problems

(a) If the grain is approximately hemispherical, and the larger capacitance C_2 is identified as that of the curved surface of the hemisphere to a surrounding metal electrode, what is the radius of the grain assuming that the oxide layer has the properties stated in paragraph (b) of Sec. III A.

(b) Under these assumptions what is the volume of the grain?

B. Grain size—answers

(a) From the specified properties of the oxide layer the capacitance per unit area is

$$\frac{\kappa\epsilon_0}{d}=\frac{8}{36\pi\times10^9\times10^{-9}}$$
$$=0.07\ \mathrm{F\,m^{-2}}=0.07\ \mathrm{aF(nm)^{-2}}.$$

Accordingly, the area is $A=8/0.07=114\ (\mathrm{nm})^2$. Equating this to the curved area of a hemisphere $2\pi R^2$, gives $R=4.3$ nm.

(b) The volume of a hemispherical grain of this radius is $(2\pi/3)R^3=166\ (\mathrm{nm})^3$.

V. COULOMB BLOCKADE AND STAIRCASE

The *Coulomb blockade* occurs because no current can flow through the grain until a threshold voltage V_{th} is reached that is sufficient to supply the energy needed to move an electron onto or off from the grain. For example, from inspection of the $I(V)$ curve in Fig. 2, we see that the blockade (where $I\approx 0$) extends from ~ -6 mV to $\sim +8$ mV, allowing for some smearing by thermal energies.

In simple circumstances the width of the Coulomb blockade can be estimated as follows: The energy cost of putting an electron on an initially neutral grain with capacitance C is $e^2/2C$. This energy can be supplied by the external voltage V when it is large enough to establish a voltage ϕ at the grain such that $e\phi \leq e^2/2C$. The width is then twice this voltage.

The term *Coulomb staircase* refers to the series of equally spaced sloping steps in which the $I(V)$ curve rises at higher voltages. The spacing of these steps is about 37 mV in the $I(V)$ curve in Fig. 2. This voltage interval is that in which the mean number of electrons on the grain changes by one.

In a real experiment, the metal grain under investigation is usually electrically polarized by random charged impurities that are nearby and electrostatically coupled to the particle.[4] Because it is produced by polarization, the resulting "offset charge" Q_o can have *any* value and, hence, is usually *not* an integer multiple of e. Therefore, because the total charge on the grain is always an integer multiple of e, the sum of the charges on C_1 and C_2 will be $(Q_o - ne)$ if n extra electrons are on the grain relative to a neutral state. If no voltage V is applied to the device, the electrostatic energy in the capacitors is simply

$$E=\frac{(Q_o-ne)^2}{2(C_1+C_2)} \tag{1}$$

since C_1 and C_2 are in parallel, seen from the grain. Thus, in the minimum energy state, n takes the integer value n_o closest to Q_o/e so that $|Q_o - n_o e| \leq e/2$.

A. Coulomb blockade and staircase—problems

(a) Suppose the grain is polarized by an offset charge Q_o and has n excess electrons. Find the change in electrostatic energy of the system when an electron is added.

(b) Find an expression for the potential ϕ (with respect to ground) presented at the midpoint of the capacitive voltage divider when the bias voltage across the device is V.

(c) If $Q_o=0$, at what threshold voltage V_{th} will the energy $e\phi$ provided by the voltage source be sufficient to supply the electrostatic energy needed to move an electron from ground to the grain, causing the electron number to go from n to $n+1$? How does the value of V_{th} for $n=0$ compare with the measured Coulomb blockade?

(d) What is the voltage spacing between successive steps in the Coulomb staircase? How does the spacing shown in Fig. 2 compare with that computed from the parameter values quoted above?

B. Coulomb blockade and staircase—answers

(a) Using Eq. (1), the energy difference is

$$\Delta E \equiv E_{\mathrm{final}}-E_{\mathrm{initial}}=\frac{[Q_o-(n+1)e]^2}{2(C_1+C_2)}-\frac{(Q_o-ne)^2}{2(C_1+C_2)}$$
$$=\frac{e^2}{2(C_1+C_2)}\left(2n+1-\frac{2Q_o}{e}\right).$$

In the simple case of $Q_o=n=0$, the energy increment is $e^2/2(C_1+C_2)$. On the other hand, notice that in the special case $Q_o/e-n=1/2$, the energy difference is *zero.* Thus fractional offset charges have a major effect on charging energies and cannot be ignored when interpreting experimental results.

(b) This is a standard problem for capacitors in series. The total capacitance is $C_t=C_1C_2/(C_1+C_2)$, and the charge Q is the same on each capacitor. The total voltage drop across the two capacitors is $V=Q/C_t$ and that across C_1 is $\phi=Q/C_1$. Eliminate Q by dividing the second equation by the first, and you get

$$\phi=\frac{C_2V}{C_1+C_2}$$

for the voltage across C_1.

(c) Equate the energy $e\phi$ from (b) with the increase in charging energy ΔE from (a), and solve for the threshold voltage $V_{\mathrm{th},1}$ for electron transfer onto the grain across C_1. For $Q_o=0$, we have

$$e\frac{C_2V}{C_1+C_2}=\frac{e^2}{2(C_1+C_2)}(2n+1)$$

or

$$V_{\mathrm{th},1}=\frac{e}{2C_2}(2n+1). \tag{2}$$

In the Coulomb blockade region, $n=0$ if $Q_o=0$, and the threshold voltage for transferring an electron across C_1 to the grain will be $e/2C_2$. The threshold for transferring an electron across C_2 will be $e/2C_1$ by the same reasoning. The observed threshold for current flow will be the lower of these two, namely, $e/2C_>$, where $C_>$ is the greater of C_1 and C_2. With the quoted values: $C_>=C_2=8$ aF; this gives $V_{\mathrm{th}}=10$ mV if $Q_o=0$. In contrast, as noted above, if Q_o were $e/2$, there would be no Coulomb blockade, and $V_{\mathrm{th}}=0$. The experimental value ~7 mV lies between these limiting cases.

(d) The successive steps in the Coulomb staircase occur at the threshold voltages for changing the average electron number from n to $(n+1)$. Applying the formula Eq. (2) derived above, you can see that the spacing between the n'th and $(n'+1)$st steps is $\Delta V=e/C_2$ if the charge transfer across C_1 dominates, or e/C_1 if transfer across C_2 dominates. From Fig. 2 a reasonable estimate of the average step spacing is 37 mV. This result yields a capacitance value of 4.3 aF, in reasonable agreement with the value $C_1=4.9$ aF determined using a computer simulation to fit the entire $I(V)$ curve.[5]

VI. Energy levels of electrons in the grain

The Coulomb blockade threshold voltage is determined in Sec. V assuming that the only relevant energy in the problem is the electrostatic energy of the extra electron on the grain. According to quantum mechanics, however, the electron also has kinetic energy from being localized in the grain, i.e., in a box.[6] The allowed energy levels are discrete with average separation ΔE between them equal to the reciprocal of the density of states $D(E)$.

Properties of such states can be estimated by modeling the mesoscopic system as a cube of edge length L as long as we also assume that there are geometric imperfections that split any exact degeneracies arising from the symmetry of a cube. The eigenenergies E of an electron with mass m in such a box are given by the well-known relation

$$E=\frac{\pi^2\hbar^2}{2mL^2}(n_x^2+n_y^2+n_z^2),$$

where $\hbar=h/2\pi$ is the reduced Planck's constant and n_x, n_y, and $n_z=1,2,3,4,\ldots$. This yields a density of states $D(E)$ such that the number of states with energy between E and $E+dE$ is

$$D(E)dE=\frac{m^{3/2}L^3}{2^{1/2}\pi^2\hbar^3}E^{1/2}dE.$$

The Fermi energy E_F is defined to be the highest energy reached when these levels are filled with N particles:

$$N=\int_0^{E_{\mathrm{F}}}2D(E)dE,$$

where the factor of 2 takes into account the fact that the two possible electronic spin states permit two electrons to be in each energy state. This will give

$$E_{\mathrm{F}}=3^{2/3}\pi^{4/3}\frac{\hbar^2}{2m}n^{2/3},$$

where n is the number of particles per unit volume.

As the bias voltage V is increased above the Coulomb blockade value, an additional resonant tunneling channel opens each time the available energy from the voltage source passes through one of the discrete energy levels, and discrete steps like those in Fig. 3(a) occur in the current. These steps are more easily visible as peaks in the plot of dI/dV in Fig. 3(b).

These discrete steps will be washed out into a smooth $I(V)$ curve by thermal energy unless k_BT is small compared with the level spacing ΔE. (Here, $k_B=1.38\times10^{-23}$ J K^{-1} is the Boltzmann constant.)

A. Energy levels—problems

(a) Given the electron density in aluminum mentioned above, find the Fermi energy E_{F}.

(b) Given the volume of the aluminum grain estimated in Sec. IV B(b), find the energy level spacing at the Fermi energy. How does this compare with the average step spacings of the data in Fig. 3? In making this comparison, remember that only the fraction $C_2/(C_1+C_2)$ of the energy eV is available, so that the observed voltage separation ΔV must be scaled down by this fraction before being compared with the level spacing ΔE.

(c) Estimate the maximum temperature at which individual electronic energy levels can be resolved in such measurements when the grain has a volume $\mathcal{V}$. What is this temperature for the grain described above? Your answer will show why data taken at 4.2 K (Fig. 2) do not show these levels, but data taken at 0.32 K (Fig. 3) do.

B. Energy levels—answers

(a) Inserting the electron density $n=1.8\times10^{23}$ cm^{-3} into the expression for E_{F} and $n=N/\mathcal{V}$ gives

$$E_{\mathrm{F}}=3^{2/3}\pi^{4/3}\frac{\hbar^2}{2m}n^{2/3}=1.85\times10^{-18}\ \mathrm{J}=11.6\ \mathrm{eV}.$$

(b) From Fig. 3 there are 8 ± 1 peaks in a range of 4.5 mV. Therefore, the average voltage difference between peaks is (4.5 mV)/(7 ± 1)=650 ± 100 μV.

From the relationship between E_{F} and $n=N/\mathcal{V}$, you can see that

$$\frac{dE_{\mathrm{F}}}{dN}=\frac{2}{3}\frac{E_{\mathrm{F}}}{N}=0.67\times\frac{11.6}{1.8\times10^{23}\times166\times10^{-21}}=260\ \mu\mathrm{eV}.$$

Note that this average value applies to individual quantum states. When the two-fold spin degeneracy is taken into account, the average spacing between the doubly degenerate levels will be 520 μeV. Correcting for the $C_2/(C_1+C_2)$ voltage divider factor leads to the expectation that the ΔV between the successive peaks will be ~780 μV if $C_2=2C_1$. This compares reasonably well with the value of 650 μV observed in Fig. 3, considering the irregularity of the spacing.

(c) To obtain an order of magnitude estimate of how cold the sample should be to exhibit discrete electron energy levels, set $k_BT=2E_{\mathrm{F}}/3n\mathcal{V}$ and solve for the temperature T in kelvins. This leads to

$$T=\frac{2}{3}\frac{E_F}{n\mathscr{V}k_B}=\frac{5\times10^{-19}}{\mathscr{V}},$$

where $\mathscr{V}$ is the volume of the grain in cubic centimeters or $500/\mathscr{V}$ if $\mathscr{V}$ is in cubic nanometers.

Inserting the volume $\mathscr{V}=166\ (\mathrm{nm})^3$ found earlier leads to the requirement that $T<3$ K in order for the discrete level structure to be observable. A more careful analysis shows that the effective broadening is $\sim4k_BT$, so that $T<0.75$ K is a better estimate for this grain. In the absence of a magnetic field strong enough to split the spin degenerate states by an amount comparable to the spacing, $T<1.5$ K is enough. Whatever the case, the result shows that even in metallic grains as small as these, the discreteness in the level structure is only observable at very low temperatures.

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[1]Konstantin K. Likharev and Tord Claeson, "Single electronics," Sci. Am. **266**(6), 80–85 (1992).

[2]Marc A. Kastner, "Artificial atoms," Phys. Today **46**(1), 24–31 (1993).

[3]D. C. Ralph, C. T. Black, and M. Tinkham, "Spectroscopic measurements of discrete electronic states in single metal particles," Phys. Rev. Lett. **74**, 3241–3244 (1995).

[4]See, for example, A. E. Hanna and M. Tinkham, "Variation of the Coulomb staircase in a two junction system by fractional electron charge," Phys. Rev. B **44**, 5919–5922 (1991).

[5]The step spacing is determined by C_1 rather than C_2 because changes in the average value n will be controlled dominantly by the threshold for the lower resistance junction, which, in turn, depends on the capacitance across the *other* junction. This argument implies that the spacing between steps will be given, in general, by

$$\Delta V=\frac{e}{C_{R>}},$$

where $C_{R>}$ denotes the value of whichever of C_1 or C_2 has the greater tunneling resistance. The parameters for the grain used here as an example are consistent with this result.

[6]The large mutual repulsive Coulomb energy of the conduction electrons is essentially canceled by their attraction to the positive cores if the grain is neutral. That is why only the *extra* electron enters in the analysis of Sec. V.

THE ENGINEERING PROFESSION

I was much gratified in receiving a letter from you some months ago informing me that you had commenced to prepare yourself in the most thorough manner for the business of practical engineering. I would have answered it immediately but was prevented at the time by a press of college duty and other engagements and as usual when I do not attend to a letter as soon as it is received I have suffered your communication to remain unanswered until a late day. Each hour brings its own duties and I have long since found by sad experience the evils of procrastination.

I think you have made a good choise and provided you can persever in the course you are now in there is every probability that you will become an important and useful man. The professions of Law and Medicine are so much crowded that provided a person has the proper talents for the business of practical engineering I think his chance of success is greater in this line than in either of the others. I think it requires more originality of mind to make a good practical engineer than a passable Lawyer or Doctor. It requires but little mind to attain a knowledge of the ordinary forms of law pleadings and the mistakes of the Physician are often buried with his patient while the labours of the engineer are seen and appreciated or at least criticised by many.

Joseph Henry, letter to a former student (1845), in *The Papers of Joseph Henry*, edited by Marc Rothenberg (Smithsonian Institution Press, Washington, 1992), Vol. 6, pp. 246–247.

CAT PHYSICS

I am glad to learn that you are still interested in subjects pertaining to your college studies. The phenomenon you mention although an interesting one is as old as the days of Pliny. If the fore finger be placed on the back and the thumb under the neck a slight shock will sometimes be felt through the hand when the cat is rubbed.

I presume you are making good progress in your study of the law...

Joseph Henry, letter to William Gledhill (1844), in *The Papers of Joseph Henry*, edited by Marc Rothenberg (Smithsonian Institution Press, Washington, 1992), Vol. 6, p. 131.