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Principle of a variable capacitor based on Coulomb blockade of nanometric-size clusters

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Abstract. – We show how Coulomb blockade of electrons in a dispersive set of clusters embedded in the dielectric of a capacitor can be used to design a voltage tunable variable capacitor (varactor). We calculate the variation of capacitance for typical size distribution of the clusters and as a function of the dielectric constants of the insulators. We also discuss the temperature and frequency dependence of the capacitor.

Introduction. – Studies of single-electron tunneling started more than thirty years ago with Zeller and Giaver [1], and Lambe and Jaklevic [2]. Nevertheless, it is at the end of the eighties that Coulomb blockade became the subject of extensive experimental [3–6] and theoretical [7–12] studies. This has led to the development of the physics of artificial atoms [13] and also to various applications in microelectronics [14]. However, most of the studies have been done on I-V or $\mathrm{d}I/\mathrm{d}V$-V spectra. Beside the early work of Lambe and Jaklevic [2] and Cavicchi and Silsbee [15], only few experiments have been performed to measure the capacitance variation due to single-electron tunneling. In addition, most of those studies were done on semiconductor quantum dots [16–19].

In this article, we study a configuration similar to the original one of Lambe and Jaklevic. We show how a progressive change of capacitance can be obtained by using a bias voltage to control the Coulomb blockade of an appropriate set of clusters. We derive the variation of the capacitance as a function of the distribution of size in the set of clusters.

Description of the model. – Let us consider the nanometer-sized structure of fig. 1, where a small nanometric-size island is separated from the left and right electrodes by, respectively, a thin tunnel junction and a thick insulating barrier. The thickness (capacitance) of the left and right insulating layers are, respectively, d_{L} (c_{L}) and d_{R} (c_{R}) with $d_{\mathrm{L}} < d_{\mathrm{R}}$. In this structure, d_{R} is chosen to be large enough to prevent tunnelling between the island and the

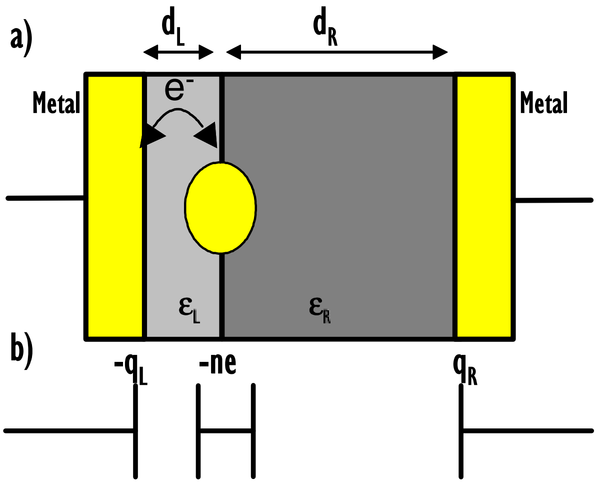

Fig. 1 – a) A nanometric metallic island is embedded in-between two insulating layers and two metallic electrodes. Electrons can tunnel from the left electrode to the island. In contrast, the thicker right insulating layer prevents tunnelling to occur on the measurement timescale. b) Schematic representation of the structure as two capacitors in series. $q_{\rm L}(q_{\rm R})$ is the charge build-up on the right electrode of the left (right) capacitor. $q_{\rm L} - q_{\rm R} = -ne$ is the charge in excess on the island.

right electrode in the measurement timescale. As tunnelling depends exponentially upon the barrier thickness, this condition is easily achieved without having $d_{\rm L} \ll d_{\rm R}$.

In the following, we will use the framework of the orthodox theory of Coulomb blockade and we will consider the case where $E_{\rm c} = e^2/2(c_{\rm L} + c_{\rm R}) \gg k_{\rm B}T$, $E_{\rm c}$ being the charging energy of the island. As electrons can only tunnel through the left junction, there is no net current flowing through the whole structure at a fixed voltage V. Nevertheless, for $V_{c_n} < V < V_{c_{n+1}}$ (where $V_{c_n} = (2n-1)e/2c_{\rm R}$ is the threshold voltage for charging a n-th electron onto the island), the island has an integer n excess of charges $-ne$.

In the presence of an applied voltage $V < V_{c_1}$, there is no excess charge on the island, we have the same charge build-up on the right and left capacitor $q_{\rm L} = q_{\rm R}$ and we obtain

$$V = \frac{q_{\rm R}}{c_{\rm R}} + \frac{q_{\rm L}}{c_{\rm L}} = \frac{c_{\rm L} + c_{\rm R}}{c_{\rm L} c_{\rm R}} q_{\rm R} = \frac{q}{c}\,, \tag{1}$$

where c is the equivalent capacitance of the global structure (limited to the island zone) and q its charge. For an applied voltage $V_{c_n} < V < V_{c_{n+1}}$, we have $q_{\rm R} = q$ for the right electrode which is electrically isolated from the island, while tunneling authorizes a distribution of the charge of the capacitance between the left electrode and the island, $q_{\rm L} = q - ne$. Equation (1) becomes

$$V = \frac{q}{c_{\rm R}} + \frac{q-ne}{c_{\rm L}} = \frac{q}{c} - \frac{ne}{c_{\rm L}} \tag{2}$$

and the charge of the capacitor is written as

$$q = cV + \frac{c}{c_{\rm L}} ne\,. \tag{3}$$

This means that q increases abruptly by $nec/c_{\rm L}$ at each step V_{c_n} of the Coulomb staircase. In other words, the differential capacitance, $c_d = {\rm d}q/{\rm d}V$ presents δ-function–like peaks at each step n and is constant ($c_d = c$) between two steps.

Let us now consider the capacitor of the type of fig. 1, but of macroscopic size in which the single metallic island is replaced by a 2D array of N metallic islands with distributed sizes. Here, S is the area of the electrodes and $D = N/S$ is the island density. For simplicity, each island will be described as a disk of surface s_i having a capacitance $c_{i,\sigma} = \epsilon_\sigma s_i/d_\sigma$, where $\sigma = \mathrm{L}, \mathrm{R}$. We call $\rho(s)$ the normalized distribution of s. The structure will be considered as N nanometer-sized junctions like the one of fig. 1 in parallel. All those nanometer-sized junctions are also in parallel with the capacitance corresponding to the zones of S without islands, that is $C_J(1-\tau) = \epsilon_\mathrm{L}\epsilon_\mathrm{R}S(1-\tau)/(\epsilon_\mathrm{R}d_\mathrm{L} + \epsilon_\mathrm{L}d_\mathrm{R})$, where τ is the island covering factor. In the following the capital letters will refer to a parameter of the whole macroscopic structure of surface S and the small letters will refer to a parameter of one of the N nanometric capacitors. We neglect mutual capacitance between islands.

The total charge of the structure, Q, is obtained by adding up the contributions from all the islands, given by eq. (3) with $n = n_i$, $c = c_i$ and $c_\mathrm{L} = c_{i,L}$ for the i-th island, and the contribution $C_J(1-\tau)$ from the zones without islands. We have

$$\begin{aligned} Q &= C_J(1-\tau)V + \sum_{i=1}^{N} c_i \left[V + \frac{n_i e}{c_{i,\mathrm{L}}} \right] && (4) \\ &= C_J V + \sum_{i=1}^{N} \left[\frac{\epsilon_\mathrm{R} d_\mathrm{L}}{\epsilon_\mathrm{R} d_\mathrm{L} + \epsilon_\mathrm{L} d_\mathrm{R}} n_i e \right]. && (5) \end{aligned}$$

When a small a.c. voltage v is added to the bias voltage V, the differential capacitance $C_d = \mathrm{d}Q/\mathrm{d}v$ can be written as

$$C_d(V) = C_J + \frac{\epsilon_\mathrm{R} d_\mathrm{L} e}{\epsilon_\mathrm{R} d_\mathrm{L} + \epsilon_\mathrm{L} d_\mathrm{R}} \left[\frac{\mathrm{d}}{\mathrm{d}v} \left(\sum_{i=1}^{N} n_i \right) \right]_V . \tag{6}$$

The islands accommodating a n-th additional electron at the potential V are those of surface

$$s(n,V) = \frac{2n-1}{2} \frac{d_\mathrm{R} e}{\epsilon_\mathrm{R} V}. \tag{7}$$

From that, the number of those islands accommodating an additional electron in the voltage range between V and $V + \mathrm{d}v$ is

$$\begin{aligned} d\left(\sum_{i=1}^{N} n_i \right) &= \sum_n N\rho(s)\mathrm{d}s && (8) \\ &= \sum_n N\rho(s(n,V)) \frac{2n-1}{2} \frac{e d_\mathrm{R}}{\epsilon_\mathrm{R} V^2} \mathrm{d}v . && (9) \end{aligned}$$

In fine, the differential capacitance can be written as

$$C_d = C_J + \frac{d_\mathrm{L} d_\mathrm{R}}{\epsilon_\mathrm{R} d_\mathrm{L} + \epsilon_\mathrm{L} d_\mathrm{R}} \sum_n \left[N\rho(s(n,V)) \frac{2n-1}{2} \frac{e^2}{V^2} \right] . \tag{10}$$

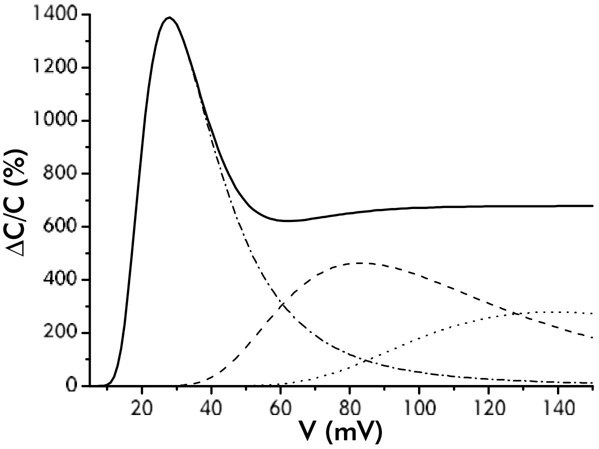

Fig. 2 – Variation of differential capacitance *vs.* d.c. voltage (solid curve) expected for a set of gold clusters having a Gaussian diameter distribution (extracted from ref. [20]) with $\overline{r} = 2.2$ nm, $\sigma = 0.5$ nm and $D = 3.99 \cdot 10^{16}/\mathrm{m}^2$. The two insulating barriers are $\mathrm{SiO_2}$ and $\mathrm{SrTiO_3}$ with respective parameters: $d_\mathrm{L} = 2.5$ nm, $d_\mathrm{R} = 5$ nm and $\epsilon_\mathrm{L} = 3.9$, $\epsilon_\mathrm{R} = 332$. The respective capacitance contributions of the first three orders of n are also given: $n = 1$ (dash-dotted curve), $n = 2$ (dashed curve) and $n = 3$ (dotted curve).

This gives for the variation ΔC of C_d between $V = 0$ and V:

$$\frac{\Delta C}{C_J} = \frac{C_d - C_J}{C_J} = \left[d_\mathrm{L} d_\mathrm{R} D \rho(s(n,V)) \frac{2n-1}{2} \frac{e^2}{\epsilon_\mathrm{L}\epsilon_\mathrm{R} V^2} \right]. \tag{11}$$

We have calculated $\Delta C/C_J$ from eq. (11) for the typical size distribution which has been found for a set of gold clusters embedded in an oxide layer (sets easy to produce by sputtering) [20]. We have supposed that these clusters are embedded between $\mathrm{SiO_2}$ (low dielectric constant, $\epsilon_\mathrm{L} = 3.9$) and $\mathrm{SrTiO_3}$ (high dielectric constant, $\epsilon_\mathrm{L} = 332$) layers. As shown in fig. 2, the variation of the differential capacitance as a function of the bias voltage reaches 1400%. The first three orders in n are detailed as a guideline as the total capacitance variation corresponds mainly to the sum of these three contributions. The variation of $\Delta C/C$ of fig. 2 is typical of a relatively narrow size distribution, with a main peak (for $n = 1$) weakly overlapping with the higher-order contributions. Sharper peaks with higher values of $\Delta C/C$ could be obtained with narrower size distributions. In fig. 3, we illustrate the opposite case for which $\Delta C/C$ is smaller but extends over a broader voltage range: in this example, this is due to a larger cluster size distribution and a lower ϵ_R.

Following eq. (11), for a Gaussian distribution of the islands surfaces $\rho(s)$ with a mean value $\overline{s}$ and a narrow standard deviation $\sigma \ll \overline{s}$, the maximum variation of differential capacitance occurs at $V \simeq ed_\mathrm{R}/2\epsilon_\mathrm{R}\overline{s}$ for $n = 1$ and is

$$\frac{\Delta C}{C_J} = \left[\frac{2D\overline{s}^2}{\sigma\sqrt{2\pi}} \frac{d_\mathrm{L}\epsilon_\mathrm{R}}{d_\mathrm{R}\epsilon_\mathrm{L}} \right]. \tag{12}$$

A lecture of eq. (12) gives in a simple physical way the pertinent parameters of the differential capacitance variation amplitude. The amplitude of the effect is raised by choosing a cluster distribution with a high density (D) and a low-size dispersion ($\overline{s}^2/\sigma$) and also by choosing two different dielectric constant ($\epsilon_\mathrm{R} \gg \epsilon_\mathrm{L}$) and enlarging $d_\mathrm{L}/d_\mathrm{R}$ while keeping $d_\mathrm{L} \lesssim d_\mathrm{R}$. The

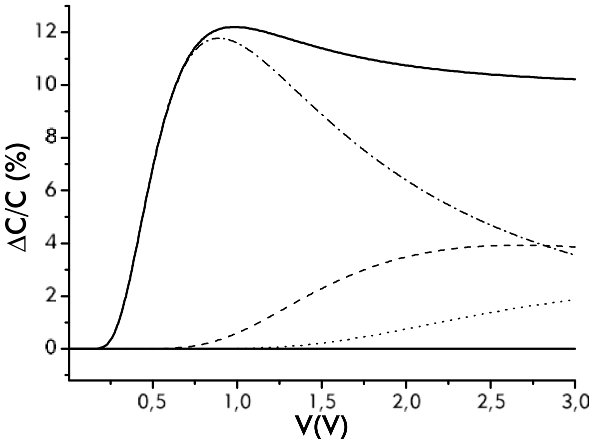

Fig. 3 – Variation of capacitance *vs.* voltage (solid curve) expected for a set of clusters having a Gaussian diameter distribution with $\overline{r} = 1.56$ nm and $\sigma = 1$ nm and $D = 3.17 \cdot 10^{16}/\mathrm{m}^2$. The two insulating barriers are SiO_2 and Al_2O_3 with respective parameters: $d_\mathrm{L} = 2.5$ nm, $d_\mathrm{R} = 5$ nm and $\epsilon_\mathrm{L} = 3.9$, $\epsilon_\mathrm{R} = 9$. The respective capacitance contributions of the first three orders of n are also given: $n = 1$ (dash-dotted curve), $n = 2$ (dashed curve) and $n = 3$ (dotted curve).

voltage range of the rising slope corresponds, approximately, to a surface variation from $\overline{s}+2\sigma$ to $\overline{s}$, so that the effective voltage control range is

$$\Delta V = \frac{e d_\mathrm{R}}{2\epsilon_\mathrm{R}} \frac{2\sigma}{\overline{s}(\overline{s}+2\sigma)} \,. \tag{13}$$

Increasing (decreasing) ϵ_R (d_R) increases the capacitance variation but decreases the voltage range. A narrower cluster size distribution has similar consequences. Capacitance variation could be increased by one order of magnitude with narrower clusters distributions [21] and by using a very high dielectric constant as thick insulator ($BaTiO_3$, $\epsilon \sim 1600$).

Frequency and temperature dependence. – The influence of frequency and temperature can be described by cut-off effects. Concerning frequency, if the alternative voltage period $1/f$ is lower than the $r_\mathrm{L} c_\mathrm{L}$ product of an island, then this island will not be able to charge-discharge during a cycle and no capacitance variation will arise from it. This gives rise to a cut-off frequency for the effect. As an example, for a 2 nm diameter cluster associated with a thin SiO_2 barrier the capacitance c_L is within the order of 10^{-19} F. To ensure the Coulomb-blockade regime, the junction resistance will have to be at least of the order of $r_\mathrm{L} = e^2/h$ giving a maximum cut-off frequency of the order of 100 THz. With respect to temperature, in order to have Coulomb-blockade effect, the cluster charging energy E_C has to satisfy $E_\mathrm{C} \gg kT$. The charging energy of a cluster in a "set of cluster" environment will be less lower than that of an isolated one. Coulomb blockade of clusters of 2 nm diameter in a "set of clusters" environment has been observed [22] at room temperature for densities close to the ones given in examples in figs. 3 and 2.

Conclusion. – In conclusion, we have described and assessed a concept of capacitor the a.c. capacitance of which can be varied by applying a bias voltage. The variation of the capacitance is governed by Coulomb-blockade effects on metallic clusters embedded in the dielectric of the capacitor. The amplitude of the capacitance variation and its bias voltage range are controlled by the size distribution of the clusters and the dielectric insulators. We have discussed the condition for obtaining a device working at room temperature and up to high frequency. We have shown that effects of interest for applied devices can be obtained with typical arrays of clusters that are currently fabricated with standard deposition techniques. Reversibly, this effect could also be used as a probe to obtain the size distribution of a set of clusters through a differential capacitance measurement.

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