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To cite this version:

Raphaelle Dianoux, Florence Marchi, Henke Jan Smilde, Nicolas Buffet, Pierre Mur, et al.. Charging dynamics and strong localization of a two-dimensional electron cloud. Nanotechnology, 2007, 18 (32), pp.325403. 10.1088/0957-4484/18/32/325403 . hal-00088811

HAL Id: hal-00088811

https://hal.science/hal-00088811v1

Submitted on 4 Aug 2006

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Charging dynamics and strong localization of a two-dimensional electron cloud

R. Dianoux,[1,2] H.J.H. Smilde,[3] F. Marchi,[4,5,*] N. Buffet,[3] P. Mur,[3] F. Comin,[1] J. Chevrier[1,4,5]

[1] *ESRF, B.P. 220, 38043 Grenoble cedex 9, France*

[2] *CEA - DRFMC/SP2M/SiNaPS, 17 avenue des Martyrs, 38054 Grenoble cedex 9, France*

[3] *CEA-DRT - LETI/DTS, 17 avenue des Martyrs, 38054 Grenoble cedex 9, France*

[4] *CNRS/LEPES, 25 avenue des Martyrs, 38042 Grenoble cedex 9, France*

[5] *Université Joseph Fourier, B.P. 53, 38041 Grenoble cedex 9, France*

ABSTRACT

The dynamics of charge injection in silicon nanocrystals embedded in a silicon dioxide matrix is studied using electrostatic force microscopy. We show that the presence of silicon-nanocrystals with a density of 10^{11} cm^{-2} is essential for strong localization of charges, and results in exceptional charge retention properties compared to nanocrystal-free SiO_2 samples. In both systems, a logarithmic dependence of the diameter of the charged area on the injection time is observed on a time scale between 0.1 and 10 s (voltage ≤ 10 V). A field-emission injection, limited by Coulomb blockade, and a lateral charge spreading due to a repulsive radial electric field are used to model this logarithmic behavior. Once the tip is retracted, the electron cloud is strongly confined in the nanocrystals and remains completely static.

PACS numbers. 07.79.Lh, 68.43.Jk, 73.23.Hk, 73.63.Bd, 81.16.Rf

[*] Electronic address: marchi@cnrs.grenoble.fr

I. INTRODUCTION

Silicon nanocrystals (Si-nc) exhibit remarkable charge retention properties in addition to excellent read/write performances, and are therefore potential candidates to replace poly-silicon in the floating gate of non-volatile memories [1-3]. This article presents experimental evidence of confinement of charges in quantum dots, using samples containing Si-nc. The confinement is very efficient, and stabilizes a dense 2D-electron cloud against strong Coulomb repulsion. Specifically, we demonstrate here that: (i) quantum dots increase the maximum density of trapped electrons by a factor of 3 compared to homogeneous silicon dioxide, and (ii) the quantum dots density is the determining parameter for electron injection, whereas in the absence of dots the local inhomogeneities of SiO_2 control the injection.

The technique employed to characterize these electric properties is electrostatic force microscopy (EFM). It is based on atomic force microscopy [4] (AFM) and allows for non-destructive investigation of the electric properties of a sample at the nanometer scale, without the need for special sample preparation. The principle consists of using an AFM to successively inject and detect charges with the tip, with a resolution in air of only a few tens of elementary charges.

First, the sample fabrication is described, and the characteristics of the nanocrystals are summarized. The technique of EFM in our experimental conditions is briefly described, along with the expected resolution. In the third section, the results of comparative experiments on the Si-nc sample and a reference SiO_2 sample are described, and emphasize in particular the differences in reproducibility, retention time and electron density. In the section thereafter, we discuss the mechanism accounting for the spreading of the charges in the Si-nc sample while the biased AFM tip is in contact, and for the static situation once the tip is removed.

II. EXPERIMENTS

To emphasize the influence of the nanocrystals on the charge retention properties, a comparative study is carried out on two samples. One contains a layer of silicon nanocrystals on top of a silicon dioxide layer on a silicon substrate, and the second serves as a reference and does not contain a nanocrystal layer (see Fig. 1 for sample geometries).

The 25-nm-thick, insulating silicon dioxide layer underneath ensures that injected charges do not leak to the grounded substrate. It is thermally grown on a (100)-oriented *p*-type silicon substrate that contains a boron impurity concentration of 10^{15} at. cm^{-3}. The nanocrystal sample is prepared by depositing a 3-nm-thick layer of silicon-rich-oxide (SRO) [5, 6] with low-pressure chemical vapor deposition (LP-CVD). Two precursor gases, SiH_4 and N_2O, are introduced in the chamber for the deposition of the SRO layer, at 515°C substrate temperature and a total gas pressure of 1000 mTorr. They react to form an amorphous SiO_x film ($x < 2$), which is then annealed during 10 min at 1000°C in nitrogen at ambient pressure. This induces a phase separation, resulting in the precipitation of crystalline silicon nanocrystals in a silica matrix. The density and

size of the Si-nc embedded in a SiO_2 matrix depend critically on the ratio of the gas fluxes used during LP-CVD. Previous high-resolution transmission electron microscopy studies have shown that Si-nc with a typical lateral dimension of 4 - 6 nm are formed, with a density between 10^{11} and 10^{12} cm^{-2} Si-nc [7]. The Si-nc sample studied here exhibits a low concentration of silicon in the SRO layer, as measured by spectroscopic ellipsometry analysis [8]. We estimate the density of nanocrystals to be of a few 10^{11} cm^{-2}.

Charging experiments are carried out in ambient temperature and pressure conditions using a commercial AFM [9].

Throughout the experiments, a permanent flux of nitrogen ensures a low relative humidity (RH < 15 %, limit of our commercial humidity detector) and avoids sample contamination and electro-chemical reactions during charging and charge evacuation paths on the surface [REF]. Under these conditions, topography modifications such as local nano-oxidation [REF], or rapid charge dissipation is not observed experimentally. As a result, reproducible experimental conditions are obtained, and this work does not address the influence of the humidity level on charge dissipation.

Rectangular, W_2C-coated tips [10] are used for sample charging and EFM characterization. The tips have spring constants below 1 N m^{-1}, and resonance frequencies between 36.8 and 52.1 kHz (measured data).

We briefly describe here the technique of electrostatic force microscopy [8, 11]. The AFM is operated in amplitude-feedback (dynamic) mode, and the cantilever is excited mechanically near its resonance frequency. For the injection process, scanning is stopped and the AFM tip is brought into contact with the sample's surface by reducing the amplitude setpoint to near zero. A voltage of 0 to −10 V is applied to the tip while the substrate is grounded. The injection time is varied between 100 ms and 10 s. For a negative tip voltage, electrons are injected, while a positive tip voltage leads to hole injection into the nanocrystals/silica. The exact injection conditions differ slightly for both polarizations, e.g. due to accumulation or depletion of the *p*-type substrate. We observed for our system that for holes, the number of injected charges is smaller than for electrons using the same absolute value of the tip voltage, corresponding to observations of Sugawara *et al.* [12]. In the present article, we limit the experiments and discussion to negative tip voltages and electron injection.

The EFM signal used is the phase of oscillation as compared to the excitation. It is recorded at a lift height of 50 nm. The resolution in charge of the experimental setup has been determined to be of a few tens of elementary charges using a plane capacitor model following Schaadt *et al.* [13] and considering thermal noise as the main limitation in the accuracy of the measurement. The lateral resolution amounts roughly to the value of the lift height used during recording of the EFM signal: here about 50 nm [14].

III. RESULTS

For both the Si-nc sample and the reference SiO_2 sample, the electron cloud is similar and exhibits a circular shape with a diameter of a few hundreds of nanometers (see Fig. 2a). The distribution of the charges presents a gaussian profile that can be attributed to the finite lateral resolution of the measurement (Fig. 2b). The reproducibility of the injection experiments at different sites, as well as the circular shape of the injected electron cloud, are in favor of a homogeneous distribution of the excess electrons at the relevant scales with respect to the tip resolution (compare with sample presenting kinetic roughening [8]).

However, a fundamental difference arises in the charge retention time: whereas charges are detected for only about 5 min on the reference sample, they are still present after several hours for the Si-nc sample. The diameter of the cloud stays constant in both cases, although the intensity of the EFM signal progressively vanishes. This observation is corroborated by Ng *et al.* [15] in the case of surface charges. We have shown previously [8] that the density of nanocrystals is crucial for the retention properties. In the present article, the Si-nc sample exhibits a low density of nanocrystals and thus the longest retention.

The circular shape of the electron cloud for both samples allows a systematic study over several experiments with the same injection conditions. To do so, we define two parameters of the cloud, as shown in Fig. 2b. The first parameter is the maximum phase shift $\Delta\phi_{max}$ of the profile. The second parameter is the diameter D of the cloud, measured at half maximum. A repetitive injection experiment at −10 V during 3 s on different positions is performed successively on both samples, and the two parameters are measured. The diameter D and the $\Delta\phi_{max}$ are reported in Fig. 3a and 3b, respectively.

We observe that the diameter of the electron cloud is systematically smaller for the Si-nc sample than for the reference SiO_2 sample, with an average value of ca. 200 nm vs. 370 nm respectively. Inversely, the maximum phase shift is systematically larger for the Si-nc sample, with an average value of 6.7° vs. 3.2° for the reference sample. Equally striking is the contrast in the reproducibility of the experiments: the Si-nc sample displays an excellent reproducibility with a standard deviation for the diameter of 8.4 nm (4.1 % of the mean value), whereas the reproducibility on the SiO_2 sample is poorer with a standard deviation of 34 nm (9.2 % of the mean). This contrast is emphasized for $\Delta\phi_{max}$, where the standard deviation amounts respectively to 0.32° (4.8 % of the mean) and 0.73° (22.8 % of the mean) for the Si-nc sample and for the reference sample.

Having observed that the electron cloud is completely static after injection, we address the question of the spreading of the charges on the surface during injection. For this study, the diameter D is measured as a function of the injection time in the range of 100 ms to 10 s. Several AFM probes (Tips 1 to 4 in Fig. 4) with varying spring constants are used, and the applied injection voltage is either −6 V or −10 V. The results are reported in Fig. 4.

In the presence of nanocrystals or not, the general behavior is the same: the increase of the diameter is slow and follows a logarithmic dependence on the injection time. The reproducibility of this logarithmic time evolution, independent from the injection voltage or used tip, implies that neither the spring constant nor the voltage (in the investigated range) play a role in the charge injection process. The logarithmic time

dependence furthermore indicates that theoretically the diameter will only reach saturation for an infinite injection time. However, the two samples differ in the initial value of the diameter (defined for an injection time of 1 s), which is, as mentioned before, smaller for the Si-nc sample (mean value: 185 nm vs. 320 nm). Moreover, the slopes are by far steeper for the SiO_2 reference sample: they exhibit a mean value of 117.5 against 22.6 for the Si-nc sample. Both the initial diameters and the slopes indicate that charges injected in the reference sample spread laterally over a larger distance. However, the standard deviations of the slopes show again a poor reproducibility of the injection experiments for the reference sample, with a standard deviation of 45 % from the mean value, vs. 16.4 % for the Si-nc sample. The spreading of the charges in SiO_2 may have a lower characteristic time but presents a poorer reproducibility.

IV. DISCUSSION

A. Density of electrons

The measurement of the electron cloud's diameter and its maximum phase shift allows an estimation of the number of injected charges. To do so, the plane capacitor model established by Schaadt *et al.* [13] is used. The expression of the detected charge q:

$$q = -\left[\{\Delta\phi\, k\, (z + (a / \varepsilon_{SiO2}))^3\, S\, \varepsilon_0\, \varepsilon_{SiO2}^2\} / \{Q\, a^2\}\right]^{1/2}, \quad (1)$$

where k is the cantilever stiffness and Q its quality factor, z is the lift height, a the oxide thickness, S the surface of the electron cloud, ε_0 the permittivity of vacuum and ε_{SiO2} the relative permittivity of silicon dioxide. The minus sign accounts for electrons. The density of the injected electrons is then: $\sigma = q / S$. To estimate this density for the Si-nc sample in the first experiment (Fig. 3), we take the average values of the diameter and of the maximum phase shift. The values of the other parameters are: $k = 1$ N m^{-1}, $Q = 100$, $z = 50$ nm, $a = 25$ nm, and $S = \pi D^2 / 4$. An electron density of ca. 3×10^{11} cm^{-2} is deduced, which is comparable to the nanocrystal density. This result is in good agreement with results reported by Guillemot *et al.* [16]. They found that the occupation level of the nanocrystals in a similar sample roughly equaled one electron per nanocrystal. In comparison, the density is about 3 times lower for the SiO_2 sample. Thus, not only is the retention time tremendously increased in the presence of nanocrystals, but also the density of the electrons is sensibly augmented.

More generally, we have determined that the electron density in the Si-nc sample always amounts at maximum to the nanocrystal density, independent from the injection time. In the following, we will assume that the electron density σ equals the nanocrystal density in the electron cloud for the Si-nc sample. The

reason why we cannot inject charges more densely is most likely due to Coulomb blockade related to the very small capacitance of the nanocrystals. Taking the expression of the capacitance of a sphere of diameter d, we find: $C = 2\pi\, \varepsilon_0\, \varepsilon_{SiO2}\, d$, which amounts to ca. 10^{-18} F for $d = 5$ nm. The charging energy e^2 / C (e is the elementary charge), which is the energy needed to add a second electron on the nanocrystal, is then equal to 160 meV. Although much smaller than the band offset between SiO_2 and Si, this characteristic energy is much higher than any other energy related to thermal excitation ($k_B\, T = 26$ meV at room temperature) or to quantum confinement. Coulomb blockade is then the determining mechanism, which drastically limits the insertion of a large number of electrons in a single nanocrystal.

B. Size of the electron cloud

Our experimental results show the existence of a saturating diameter of the electron cloud during injection. The potential difference between the sample surface and the tip in such a problem is clearly a crucial parameter. On the basis of elementary electrostatics, the electron cloud can be modeled as a charged disk with diameter D. Assuming a constant surface charge density, the potential difference ΔV between the center of the disk and tip during injection, can be expressed as:

$$\Delta V = V_{tip} - V_{SiO2} - (\sigma\, D / 4\, \varepsilon_0\, \varepsilon_{SiO2}), \qquad (2)$$

where V_{SiO2} is the initial potential at the surface of the 25 nm of oxide without Si-nc charging. At the onset of injection, when $D = 0$, ΔV amounts to ca. -1 V for the Si-nc sample, assuming a homogeneous voltage drop across the Si-nc and the SiO_2 layer. Equation (2) determines the maximum extension of the electron cloud, which is reached for $\Delta V \approx 0$, i.e. when the injected charges compensate the tip potential. The maximum diameter is $D_{max} = (4\, \varepsilon\, \varepsilon_{SiO2} / \sigma)\, \{V_{tip} - V_{SiO2}\}$. For an applied voltage $V_{tip} = -10$ V and a nanocrystal density of 3×10^{11} cm^{-2}, it follows that $D_{max} = 300$ nm. Although this diameter estimate comes out of the most simple analysis, it appears from Figs. 3 and 4 that experimentally determined diameters are not very far from this limit in the case of the Si-nc sample. The injected charges create a surface potential that screens the tip voltage and suppresses field emission. Injection is gradually slowed down due to Coulomb blockade, which experimentally prohibits the presence of more than one electron per nanocrystal.

In the case of the reference sample, the lower electron density implies that the diameter of the electron cloud be larger to compensate the potential difference with the tip. We determined that the electron density was 3 times lower; therefore D_{max} amounts to ca. 900 nm. Experimentally, we measure a diameter of up to 700 nm, which means that we have not yet reached electrostatic equilibrium.

C. Role of the Si-nc

From the comparison of the electron density and the dimension of the cloud between the two types of samples, it is clear that the presence of Si-nc induces strong charge localization. To understand the transport mechanism and the retention time of the electrons in the Si-nc sample, let us compare the energy diagrams of both samples (see Fig. 5). The voltage applied between the AFM probe and the substrate produces a sufficiently high electric field, on the order of 10^7 V m^{-1}, to inject charges through field emission into the conduction band of SiO_2. The electrons can then reach energetically more favorable sites, such as impurity sites (concentration 10^{15} cm^{-3}) or defects of SiO_2 (Fig. 5a). We observe that the electrons spread away from the injection site. Whether they tunnel to other impurity sites or diffuse along the surface of SiO_2 cannot be inferred from our experiments. However, based on their recent experiments, Lambert *et al.* [17] come to the conclusion that the charges disperse in SiO_2 through the traps. The fact that we do not observe an increase of the diameter of the electron cloud with increasing time after the tip is retracted, combined with a decreasing EFM signal, i.e. a slow disappearance of the electron cloud, is an indication that the electrons probably move into the volume of the silicon oxide until they reach the substrate. This situation is experimentally found to be modified by the presence of nanocrystals characterized by deep-lying energy levels, which may trap the electrons (Fig.5b). The low density of the nanocrystals (average spacing between two Si-nc: 10 nm) combined with the high potential barrier separating the conduction bands of Si and SiO_2 (3.2 eV, see Ref. [18]) reduce the tunneling probability to practically null, once the charges are trapped into the Si-nc.

Indeed, let us evaluate the tunneling rate Γ of the electrons between neighboring nanocrystals during injection. While the biased tip is in contact with the sample, a flow of electrons saturates all the available nanocrystals beneath the tip. Simultaneously, the trapped electrons create a radial electric field which tends to the explosion of the electron cloud (see for ex. Ref. [19] for theoretical evidence for Coulomb explosion). Considering our experimental conditions, i.e. an electron cloud of the order of 300 nm in diameter and an inter-crystal spacing of 10 nm, the radial electric field at the border of the cloud can be estimated to approximately 10^7 V m^{-1}. That means that the voltage drop V_{nc} across two consecutive nanocrystals is on the order of 0.1 V. The tunneling current is defined by $I_t = e\,\Gamma$ and the tunneling conductance by $G_t = I_t / V$.

G_t is also expressed as $(e^2 / h)\,T$, where e^2 / h the Landauer's single-mode quantum conductance and T the transmission coefficient. For small T, the transmission through a rectangular barrier expresses as a function of the system parameters:

$$T = (16\,E / U_0)\exp(-2\,K\,W), \tag{3}$$

where U_0 is the potential well of the nanocrystal, E is the first energy level of the Si-nc, W is the distance separating two Si-nc and K is the wave vector in the oxide: $K = (2\pi / h)\sqrt{\{2\,m_{SiO2}(U_0 - E)\}}$. m_{SiO2} is the electron effective mass in SiO_2. Taking $U_0 = 3.2$ eV, $E \approx 1/10\,U_0$, $W = 10$ nm and $m_{SiO2} = 0.32\,m_0$ [18], we calculate that $T \approx 10^{-45}$. Hence, $\Gamma = e\,T\,V / h$ amounts to some 10^{-32} s^{-1}. This means that the characteristic tunneling time ($= \Gamma^{-1}$) is so huge that an electron, once trapped in a nanocrystal, will not be able to tunnel to

an empty neighboring nanocrystal despite the existence of a strong radial electric field. In conclusion, nanocrystals behave as static traps for electrons. Our experimental observations can therefore be cast into the simple scenario that, once a single electron is trapped into a Si-nc, it will never move out (on the time scale of injection) and the trap is full and saturated.

D. Evolution with the injection time

The only situation in the Si-nc sample in which the electrons propagate away from the injection site is during the injection itself. Once the injection is stopped, the electron cloud becomes static. The proposed charge injection mechanism involves a field emission type of injection that is screened progressively with the saturation with electrons of Si-nc underlying the tip.

During the injection phase, the 25-nm-thick layer of SiO_2 below the nanocrystals prohibits the direct passage from the tip to the substrate. All charges are trapped: either in the nanocrystals or the SiO_2. The number of injected electrons q is related to the diameter D of the electron cloud by the expression: $q = (\pi / 4)\, D^2\, \sigma$, thus the current I flowing from the tip to the sample during the injection stage expresses as:

$$I = \mathrm{d}q / \mathrm{d}t = (\pi / 2)\, D\, (\partial \mathrm{D} / \partial \mathrm{t})\, \sigma. \tag{4}$$

Given the strong electric field that is applied during injection, it is reasonable to assume that a field emission regime takes place. However, considering that injection takes place in the tunneling regime would essentially yield the same qualitative conclusions. According to the expression from Fowler-Nordheim [18], the current is:

$$I_{\mathrm{FN}} \propto \Delta V^2 \exp\left(-\alpha / |\Delta V|\right) \tag{5}$$

where ΔV is defined in Eq. (2), and α is a positive constant. Combining Eqs. (4) and (5), the differential equation governing D can be retrieved:

$$\mathrm{d}D / \mathrm{d}t \propto (2 / \pi\, \sigma\, D)\, \Delta V^2 \exp\left(-\alpha / |\Delta V|\right) \tag{6}$$

This equation does not provide a straightforward evolution of D versus the injection time. However, the asymptotical behaviors at short and long injection times can be evaluated. In the first moments of injection, when D is still small, $\mathrm{d}D / \mathrm{d}t$ goes to a function proportional to $1/D$, so that $D\, t_{\mathrm{inj}}$ is proportional to $\sqrt{t_{\mathrm{inj}}}$. We have not observed this behavior and assume that it takes place on a time scale shorter than the one investigated, i.e. < 0.1 s. On the other extreme, Eq. (6) presents an asymptote when ΔV goes to 0 (or equivalently when D goes to D_{max}, in this case $\mathrm{d}D / \mathrm{d}t$ goes to 0. This limit, corresponding to saturation of

the charged area, is very much consistent with the experimentally observed logarithmic dependence on the injection time. Instead of a Fowler-Nordheim regime of injection, the use of a tunneling regime with a current proportional to the voltage drop would lead to the same limiting behavior of dD / dt.

E. Reproducibility

The last point we would like to address is the better reproducibility of the injection experiments in the presence of Si-nc. As it is observed in Figs. 3 and 4, the standard deviations of the parameters D and $D\,\phi_{max}$ are systematically higher for the reference sample. This is a clear experimental observation and therefore a characteristic behavior in these experiments. Our analysis of the system and experimental conditions lead us to the conclusion that two factors are certainly at the origin of the low reproducibility in the reference sample: first, the nanoscale inhomogeneities that could affect the mobility of the electrons in the oxide. Second, the quality of the tip-sample contact during injection is low because the oscillation of the cantilever is not interrupted during injection. Therefore, the contact is not well controlled and the tip-sample tunneling conductance is not constant over all the experiments. In order to simply model this situation and extract the key parameter, we first consider that for the long time scale, the injection is of the tunneling type, which implies that the current I is proportional to the potential difference ΔV through the tunneling conductance G_t: $I = G_t\,\Delta V$. This expression, combined with Eqs. (4) and (2) provide the evolution of the diameter when it is very close to its saturation (long time regime):

$$D = D_{max}\,\{1 - \exp(-t / \tau)\}, \tag{7}$$

where $\tau = 2\,\pi\,\varepsilon_0\,\varepsilon_{SiO2}\,D_{max} / G_t$ is a parameter dependent on G_t and σ, whereas D_{max} is only dependent on σ. In the reference sample, a local change of the electron density σ induces a change in D_{max} as well as in τ. On the other hand, the homogeneous presence of nanocrystals locks the value of the electron density and therefore makes the experiments reproducible by fixing D_{max} and τ. Therefore, we conclude that the inhomogeneities in SiO_2 are responsible for the low reproducibility in the reference sample, and not the tunneling conductance controlled by the tip-sample contact quality. The fact that the tip-sample contact is not the key parameter makes sense because the contact is essentially the same in both cases for two main reasons: (1) all the AFM probes used in our experiments are issue from the same model and manufacturer (i.e. they present similar spring constants and tip radii), (2) the surface roughness is similar for the two studied samples (less than 5 nm).

V. CONCLUSION

Using electrostatic force microscopy, we have compared the charge injection and retention properties of two different types of samples. The first contains a low density of silicon nanocrystals embedded in silicon dioxide, and the second contains only a thin SiO_2 layer and serves as a reference sample. We have shown that in the presence of nanocrystals: (i) the retention time is significantly increased as compared to the reference sample and amounts to hours vs. minutes, and (ii) the density of stored electrons is tripled, and equals roughly to one electron per nanocrystal. The electrons are trapped lastingly in the nanocrystals because of the large energy gap difference between silicon and silicon dioxide (ca. 3.2 eV). Due to the small size of the nanocrystals, Coulomb blockade is strong and prohibits the presence of more than one electron per Si-nc for the relevant energy scales. However, as soon as injection is stopped, the strong radial electric field that initially allowed the electrons to spread laterally in the oxide reduces rapidly. Hence, the situation becomes static and the electrons are trapped in the nanocrystals without a possibility to tunnel to a neighboring nanocrystal. The exceptional charge retention properties of such Si-nc samples are of interest for the application in Si-nanocrystal non-volatile memories.

Acknowledgments

This work has been carried out in the frame of the French national project ACI NanoSciences ‘Sub-PicoNewton’, and in the frame of CEA-LETI / CPMA collaboration, with PLATO Organization teams and tools. The authors acknowledge fruitful discussions with J.P. Julien who carried out electrostatic calculations.

References

[1] S. Tiwari, F. Rana, H. Hanafi, A. Hartstein, E. F. Crabbe, and K. Chan, Appl. Phys. Lett. **68**, 1377 (1996).

[2] A. Kohno, H. Murakami, M. Ikeda, S. Miyazaki, and M. Hirose, Jpn. J. Appl. Phys. **40**, L721 (2001).

[3] S. Banerjee, S. Huang, T. Yamanaka, and S. Oda, J. Vac. Sci. Technol. B **20**, 1135 (2002).

[4] G. Binnig, C. F. Quate, and C. Gerber, Phys. Rev. Lett. **56**, 930 (1986).

[5] M. L. Hitchman and J. Kane, J. Cryst. Growth **55**, 485 (1981).

[6] W. Y. Ching, Phys. Rev. B **26**, 6610 (1982).

[7] N. Buffet, P. Mur, B. D. Salvo, and M. N. Séméria, Proceedings of the IEEE Conference on Nanotechnology, 26-28 August (2002), Washington DC, USA, p. 269-272.

[8] R. Dianoux, H. J. H. Smilde, F. Marchi, N. Buffet, P. Mur, F. Comin, and J. Chevrier, Phys. Rev. B **71**, 125303 (2005).

[9] Dimension 3100, Nanoscope III AFM from Veeco Instruments, Santa Barbara, California (USA).

[10] MikroMasch, Tallinn, Estonia.

[11] M. W. Nelson, P. G. Schroeder, R. Schlaf, and B. A. Parkinson, J. Vac. Sci. Technol. B **17**, 1354 (1999).

[12] Y. Sugawara, Y. Fukano, T. Uchihashi, T. Okusako, S. Morita, Y. Yamanishi, T. Oasa, and T. Okada, J. Vac. Sci. Technol. B **12**, 1627 (1994).

[13] D. M. Schaadt, E. T. Yu, S. Sankar, and A. E. Berkowitz, Appl. Phys. Lett. **74**, 472 (1999).

[14] R. Dianoux, F. Martins, F. Marchi, C. Alandi, F. Comin, and J. Chevrier, Phys. Rev. B **68**, 045403 (2003).

[15] C. Ng, T. Chen, M. Tse, V. Lim, and A. Tseng, Appl. Phys. Lett. **86**, 152110 (2005).

[16] C. Guillemot, P. Budeau, J. Chevrier, F. Marchi, F. Comin, C. Alandi, F. Bertin, N. Buffet, C. Wyon, and P. Mur, Europh. Lett. **59**, 566 (2002).

[17] J. Lambert, G. de Loubens, C. Guthmann, M. Saint-Jean, and T. Mélin, Phys. Rev. B **71**, 155418 (2005).

[18] M. Depas, B. Vermeire, P. W. Mertens, R. van Meirhaeghe, and M. Heyns, Sol. State Electronics **38**, 1465 (1995).

[19] H.-P. Cheng and J. D. Gillaspy, Phys. Rev. B **55**, 2628 (1997).

[20] A. Haug, Theoretical and solid state physics, vol. I (Pergamon Press, Oxford, 1972).

Figure Captions

Fig. 1. (Color online) Geometries of the two samples. a) Reference, SiO_2 sample. b) Sample containing silicon nanocrystals.

Fig. 2. (Color online) a) $1 \times 0.5\ \mu m^2$ EFM image of injected charges on the nanocrystal sample. The darker area indicates the presence of excess electrons. b) Profile across the electron cloud, characterized by its maximum phase shift $\Delta\phi_{max}$ and diameter D (full-width-at-half-maximum).

Fig. 3. (Color online) Repeated injection experiments of –10 V / 3 s on different positions of the Si-nc sample and the reference sample. a) Measured diameter D. b) Measured maximum phase shift $\Delta\phi_{max}$.

Fig. 4. (Color online) Evolution of the diameter of the electron cloud with the injection time. The symbols are the experimental data points and the lines are the least-square fits. a) Injection experiments performed on the Si-nc sample. b) Injection experiments performed on the reference SiO_2 sample.

Fig. 5. (Color online) Energy diagrams of the 2 samples. a) In the SiO_2 sample, the injected electrons are trapped into the energetically low-lying but densely concentrated sites. b) In the Si-nc sample, electrons eventually reach the energetically deep-lying nanocrystals where they are trapped lastingly.

FIG. 1.

FIG. 2.

FIG. 3.

FIG. 4.

FIG. 5.