Contents lists available at ScienceDirect



Materials Science and Engineering B



journal homepage: www.elsevier.com/locate/mseb

Topographic analysis of silicon nanoparticles-based electroluminescent devices

A. Morales-Sánchez^{a,*}, J. Barreto^b, C. Domínguez^b, M. Aceves^a, K.M. Leyva^a, J.A. Luna-López^c, J. Carrillo^c, J. Pedraza^a

^a INAOE, Electronics Department, Apartado 51, Puebla 72000, Mexico

^b Instituto de Microelectrónica de Barcelona, IMB-CNM (CSIC), Barcelona, Spain

^c CIDS-BUAP, Apdo. 1651, Puebla 72000, Mexico

ARTICLE INFO

Article history: Received 31 August 2009 Received in revised form 11 March 2010 Accepted 14 March 2010

Keywords: Silicon rich oxide Silicon nanoparticles Metal-oxide semiconductor Conductive paths Electroluminescence

1. Introduction

Intense and stable photoluminescence (PL) observed in dielectric materials containing silicon nanoparticles (Si-nps) has attracted a new effective way of obtaining silicon-based light sources [1–5]. Such characteristic opens the route toward the integration of optoelectronic functions on silicon. Nevertheless, among the different techniques to fabricate nanostructured materials. compatible ones with complementary metal-oxide semiconductor (CMOS) technology must be used. Low pressure chemical vapor deposition (LPCVD) allows obtaining silicon rich oxide (SRO) layers using oxidant species like nitrous oxide (N2O) and silicon compounds (SiH₄) as reactant gasses. SRO layers with controllable and uniform silicon excess are easily deposited by LPCVD. Silicon excess is easily controlled by changing the partial pressure ratio (Ro) between nitrous oxide (N₂O) and silane (SiH₄). Silicon excess as high as 17 at.% is obtained with Ro = 3, and stoichiometric oxide can be obtained for Ro = 100[6-8]. Comparative studies have shown that SRO films deposited by LPCVD emit a stronger PL than the ones deposited by PECVD and Si implanted SiO₂ films [9,10]. Moreover, SRO films deposited by LPCVD (SRO-LPCVD) with low silicon excess have shown more intense photoluminescence (PL) than that with high silicon excess [8,9,11]. The strongest PL is obtained with silicon excess of about 4 and 5 at.%. Spectroscopical analysis has revealed

amorales@cnyn.unam.mx, amorales7901@yahoo.com.mx (A. Morales-Sánchez).

ABSTRACT

Electroluminescent properties of silicon nanoparticles embedded in MOS devices have been studied. Silicon rich oxide (SRO) films with 4 at.% of silicon excess were used as active layers. Intense and stable light emission is observed with the naked eye as shining spots at the surface of devices. AFM measurements on these devices exhibit a remarkably granular surface where the EL spots are observed. The EL measurements show a broad visible spectrum with various peaks between 420 and 870 nm. These EL spots are related with charge injection through conductive paths created by adjacent Si-nps within the SRO. © 2010 Elsevier B.V. All rights reserved.

the presence of amorphous Si-nps embedded in SRO films with low silicon excess [8].

Using the similar SRO films in MOS-like devices, electroluminescence (EL) has been reported [4,5,12]. Nevertheless, a fast degradation of the active layer and charge trapping effects have been reported in luminescent devices producing low device endurance [5,12]. Some other authors have reported a stable EL from single silicon nanocrystals, where light emission is observed as bright spots on the structures [13]. However, these devices show only one or less than a dozen spots and with low intensity.

In this article, electroluminescence of SRO films with low silicon excess deposited by LPCVD using Poly-Si/SRO/Si devices is reported. SRO films were thermally annealed at 1100 °C for 3 h in nitrogen atmosphere in order to form Si-nps. Shining spots with intense and stable light emission are observed with the naked eye. Atomic force microscopy (AFM) measurements show a granular surface where the EL spots are observed. Although AFM measurements were done on the poly, the granular surface could be associated with surface Si-nps observed in SRO films as previously reported [11]. The EL measurements show a broad visible spectrum with various peaks between 420 and 870 nm. These EL spots are related with charge injection through conductive paths created by adjacent Si-nps within the SRO.

2. Experiment

Devices were fabricated by depositing \sim 50 nm thick SRO films with 4 at.% of silicon excess on a p-type low resistivity (0.1–1.4 Ω cm) silicon (100) substrate by LPCVD. After deposition,

^{*} Corresponding author. Tel.: +52 222 2663100; fax: +52 222 2470517. *E-mail addresses:* amorales@inaoep.mx,

^{0921-5107/\$ -} see front matter © 2010 Elsevier B.V. All rights reserved. doi:10.1016/j.mseb.2010.03.030



Fig. 1. Typical *I–V* curve measured from the MOS-like devices. Insets show images of devices when are in OFF and ON state.

SRO films were thermally annealed at 1100 °C for 3 h in nitrogen atmosphere in order to induce a silicon agglomeration. A 350 nm thick n⁺ polycrystalline silicon (Poly) gate was deposited onto the SRO film surface with area of 9.604×10^{-3} cm², which is optically semitransparent in the visible rage [14]. The backside contact was formed with 1 μ m thick Al/Cu layer by sputtering. More experimental details are given elsewhere [15,16].

The surface morphology of devices was studied using a Nanosurf EasyScan AFM system Version 2.3, operated in contact mode. A 450- μ m-long single-crystal Si cantilever operated at 12 kHz (type Vista Probes CL Contact Mode AFM Probes) was used. AFM images were analyzed using scanning probe image processor (SPIP) software [17].

The EL spectra were measured by biasing the devices with a constant DC voltage. Light emission was collected with an optical fiber, which was located right onto the surface of devices and connected to a Fluoromax 3 spectrofluorometer from Horiba Jobin Yvon controlled with a computer. Current–voltage (I-V) measurements were done with a HP4156A semiconductor parameter analyzer. At least 15 different devices were characterized exhibiting similar characteristics.

3. Results and discussion

Fig. 1 shows the typical *I–V* curve at logarithmic scale of fabricated devices. A high conduction regime was measured even for low gate voltages. The current increases rapidly from 10^{-11} to 10^{-6} A from 0 to 9 V. This behavior could be related with the characteristics of the SRO film. As known, the SRO films contain silicon excess which agglomerate after the thermal annealing forming Si-nps [8]. These Si-nps can act as nodes enhancing the high current conduction in these devices. The conduction mechanisms are discussed in further detail somewhere else [3,15,16].

When the voltage reaches ~ 10 V the current increases with a different slope and devices start to emit light. This EL emission is very stable and is observed with the naked eye as discrete shining points on the surface of the devices. Insets in Fig. 1 show images of devices when are in OFF and ON state (electric field of 4.6 MV/cm). These EL spots only appear when devices are under reverse bias (RB, positive gate voltage with respect to the substrate).

Fig. 2(a) exhibits the EL spectra of luminescent devices at different electric fields. As can be seen, EL measurements show a broad visible spectrum with various peaks between 420 and 870 nm. This behavior of several peaks has been already related to the transmittance spectrum of the top polycrystalline silicon electrode [14].

Fig. 2(b) shows the EL peak intensity at \sim 650 nm as a function of the electric field (*E*). EL intensity depends on the applied electric field. The EL becomes more intense as the electric field increases. Moreover, electroluminescent spots are obtained with a low threshold electric field of \sim 1.9 MV/cm.

Analysis of surface roughness on devices made by AFM demonstrates some peaks where the EL spots are observed, as shown in Fig. 3(a) and (b). On the other hand, there are no peaks in zones where EL is not observed, see Fig. 3(a) and (c). Although this AFM analysis is done on the polysilicon surface, the roughness could be directly related with the roughness of SRO films. In fact, a morphological analysis of surface in SRO films deposited by LPCVD has shown that the SRO films exhibit a granular surface [11]. This roughness was associated with silicon compounds clusters, Si-nps or agglomerates of Si-nps for SRO films with silicon excess similar to our films. Therefore, these silicon compounds clusters or agglomerates of Si-nps could be affecting the surface of the polysilicon exhibiting some roughness.

On the other hand, Si-nps within the SRO films can be randomly placed in such way that they create conductive paths. Under this assumption, the tunnelling current does not flow uniformly



Fig. 2. (a) EL spectra of luminescent spots at different electric fields and (b) EL intensity and current density as a function of electric field.



Fig. 3. (a) Image of an electroluminescent device and 3D AFM images- of devices where (b) EL spots are observed and (c) EL spots are not observed.

through the whole capacitor area, but instead, it passes through discrete conductive paths within the oxide, as shown in Fig. 4. An increase of the total current will result in an increase of the current density in each conductive path, which results therefore in a higher amount of radiative recombination events. Indeed, conductive paths could be connected with the surface silicon compounds clusters, Si-nps or agglomerates of Si-nps observed with AFM. Therefore, multiple conductive paths would produce multiple EL spots on the surface of devices and the EL emission would be observed with the naked eye, as shown in the image of Fig. 3(a).

When the gate is positively biased respect to the substrate, electrons are attracted to the silicon surface creating an inversion layer. Electrons from this inversion layer and holes from the Poly gate are injected toward the Poly/SRO and SRO/Si interfaces through conductive paths, respectively. As a result, emission occurs after recombination within the SRO layer. As mentioned before, it has been reported that the carriers do not flow uniformly through the whole capacitor area, but they pass through discrete conductive paths within the SRO [3,15,16] and emit at their nodes. Therefore, the light is observed as single shining spots on the surface of the MOS structure. As the electric field (gate voltage) is increased, the current (carrier injection) and then the EL intensity increase. Endurance measurements of these devices were done. The contin-



Fig. 4. Model of conductive paths formed by adjacent Si-nps within the SRO film.

uous luminescence remains stable at the same value after 15 h of continuous operation.

4. Conclusion

Multiple EL spots observed with the naked eye were obtained with MOS-like structures using a SRO film with 4 at.% of silicon excess as dielectric layer. Broad electroluminescence emission spectra with various peaks were measured. This emission was related to the recombination of charge moving through conductive paths created by adjacent Si-nps within the SRO film. AFM measurements exhibited the presence of some peaks on the device's surface on zones where the EL spots appeared. Contrarily zones where no EL spots were observed the surface was smooth. Due to the high applied electric field, electrons and holes are injected from the substrate and the poly gate, respectively, resulting in radiative recombination at the luminescent nodes. These devices show a stable and intense EL for long hours of continuous operation.

Acknowledgements

Alfredo Morales acknowledges the support received from CONACYT trough the repatriation program. This work has been partially supported by the project TEC2006-13907/MIC, financed by the Spanish Ministry of Education and Science and P2005MX03 financed by the CSIC/CONACYT.

References

- J. Barreto, M. Perálvarez, J.A. Rodríguez, A. Morales, M. Riera, M. López, B. Garrido, L. Lechuga, C. Domínguez, Physica E 38 (2007) 193–196.
- [2] M. Perálvarez, J. Carreras, J. Barreto, A. Morales, C. Domínguez, B. Garrido, Appl. Phys. Lett. 92 (2008), 241104 1–3.
- [3] A. Morales-Sánchez, J. Barreto, C. Domínguez, M. Aceves, J.A. Luna-López, M. Perálvarez, B. Garrido, Nanotechnology 21 (2010), 085710 1–5.
- [4] R.J. Walters, J. Carreras, T. Feng, L.D. Bell, H.A. Atwater, IEEE J. Select. Top. Quant. Elect. 12 (6) (2006) 1647–1656.
- [5] A. Irrera, F. Iacona, I. Crupi, C.D. Presti, G. Franzò, C. Bongiorno, D. Sanfilippo, G. Di Stefano, A. Piana, P.G. Fallica, A. Canino, F. Priolo, Nanotechnology 17 (2006) 1428–1436.
- [6] D.J. DiMaria, J.R. Kirtley, E.J. Pakulis, D.W. Dong, T.S. Kuan, F.L. Pesavento, T.N. Theis, J.A. Cutro, S.D. Brorson, J. Appl. Phys. 56 (2) (1984) 401–416.
- [7] M. Aceves, C. Falcony, A. Reynoso, W. Calleja, A. Torres, Solid-State Electron. 39 (1996) 637–644.
- [8] A. Morales, C. Domínguez, J. Barreto, M. Riera, M. Aceves, J.A. Luna, Z. Yu, R. Kiebach, Rev. Mex. Fís. S 53 (2007) 279–282.

- [9] A. Morales, J. Barreto, C. Domínguez, M. Riera, M. Aceves, J. Carrillo, Physica E 38 (2007) 54–58.
- [10] F. Flores, M. Aceves, J. Carrillo, C. Domínguez, C. Falcony, Superficies y Vacío 18 (2005) 7–13.
- [11] J.A. Luna-López, A. Morales-Sánchez, M. Aceves-Mijares, Z. Yu, C. Domínguez, J. Vac. Sci. Technol. A 27 (1) (2009) 57–62.
- [12] T. Gebel, L. Rebohle, J. Sun, W. Skopura, A.N. Nazarov, I. Osiyuk, Physica E 16 (2003) 499–504.
- [13] J. Valenta, N. Lalic, J. Linnros, Appl. Phys. Lett. 84 (9) (2004) 1459-1461.
- [14] M. Perálvarez, J. Barreto, J. Carreras, D. Navarro-Urrios, Y. Lebour, A. Morales, C. Domínguez, B. Garrido, Nanotechnology 20 (2009), 405201 1–10.
- [15] A. Morales-Sánchez, J. Barreto, C. Domínguez, M. Aceves, J.A. Luna-López, Nanotechnology 20 (2009), 045201 1–7.
- [16] A. Morales-Sánchez, Correlation between optical and electrical properties of materials containing nanoparticles, PhD thesis, Chapter 5 (2008) ISBN 978-84-691-9983-1.
- [17] J.F. Jøgensen, The Scanning Probe Image Processor (SPIP), Denmark, 2002, www.imagemet.com.