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Fabrication And Characterization of Silicon Nanocrystals Based Low-Cost Schottkey Junction Photovoltaic For Harvesting Solar Energy

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Abstract: *Solution-processed semiconductors are seen as a promising route to reducing the cost of the photovoltaic device manufacture. A photovoltaic cell can be developed from the Schottky junction between a semiconductor and a metal, with or without an insulating layer between them. This work reporting a single-layer Schottky photovoltaic device that was fabricated by drop-casting intrinsic silicon nano-crystals (Si-NCs) from colloidal solution. Here the colloidal solution of silicon nano-crystals (Si-NCs) in dichlorobenzene was prepared by ultrasonic vibration (sonication) at different sonication time. Higher ultrasonic power produced smaller Si-NCs. An optoelectronic Schottky junction solar cell with Aluminium/Silicon Nano-crystals/Indium-Tin-Oxide structure on glass substrate has been investigated with and without lanthanum fluoride insulating layer. The effect of photoactive layer (Si-NCs) thickness on absorption and photo-luminance has been studied. The surface morphology of Si-NCs layer was investigated by Scanning Electron Microscope (SEM). Capacitance-voltage (C-V) study of the Schottky device revealed that resonant tunneling of electron and charge storage was there. The conductance-voltage (G-V) study also revealed the same hysteresis. The current-voltage (I-V) characterization has been studied with and without LaF_3 at different parameters. The I-V characteristics showed a dependency on the incident light intensity that indicates its possible use in low-cost solar cell fabrication.*
Keywords: *Deposition, Photovoltaic, Schottky junction, Silicon nano-crystal, Solar cells.*

I. Introduction

The sun supplies us a clean and unlimited resource of energy, helping us relieve the energy crises and world pollution. The importance of developing efficient solar cells is obvious. Historically, conventional solar cells were built from inorganic materials such as silicon. Although the efficiency of such conventional solar cells is high, very expensive materials and energy intensive processing techniques are required. This commercial solar cells remain expensive compared to fossil fuels, around 25–50 cents per kilowatt hour in the USA [1]. Current solar cells are expected to last for 20–30 years at temperatures between −20°C and 90°C, which organic solar cells may not be able to achieve [2]. The highest carrier mobility in polymer-based organic solar cells is typically on the order of 0.1 cm^2/Vs [3-4]. Compared to inorganic semiconductors, carrier mobility in organic solar cells is still low. New ways of manufacturing solar cells that can scale up to large volumes and low cost are required. One of the inorganic solar cell is silicon nanocrystals based schottky junction solar cell that can be alternative for conventional silicon solar cell. The nanocrystals provide tunable broadband absorption and the photo stability of traditional photovoltaic materials. Lead selenide (PbSe) [5] and lead sulfide (PbS) [6] nanocrystals were used in Schottky junction solar cells. These devices are infrared harvesting and can utilize the broad solar spectrum. Silicon is an abundant and non-toxic material, and it is also well studied in industrial applications. If this conventional material can be used in a novel way (e.g., nanocrystal solution processes), the manufacturing cost of silicon-based solar cells may be reduced. The efficiency of such type of solar cell can be increased by using buffer layer.

In this work a thin layer of Indium-Tin-Oxide (ITO) was fabricated on the glass substrate using electron beam evaporation technique. After that a layer of Si-NCs was developed on that ITO coated glass by drop casting deposition of Si-NCs and dicholobenzen (DCB) mixture. Then a thin layer of aluminium (Al) was developed over the Si-NCs layer by using electron beam evaporation technique. The Al layer was used as cathode and ITO layer was used as the transparent anode. The effect of photoactive layer thickness on the absorption was studied by using U-V spectrometer. LaF_3 was also studied as buffer layer of the fabricated schottky junction solar cell. The optimum layer thickness of LaF_3 was found up to 5 nm.

II. Experimental

The experimental procedures to fabricate and to characterize the fabricated solar cells are divided in following steps

2.1 Silicon nano-crystal processing

Silicon Nanocrystals can be fabricated through a variety of techniques including ion implantation [7–8], aerosol synthesis [9–10], ion beam co-sputtering [11–12], chemical vapor deposition [13–14], and reactive evaporation of silicon-rich oxides [15], laser ablation and nanoporous silicon. In this work silicon nano-crystals were fabricated using electrochemical etching of silicon wafer [16].

2.2 Cleaning of the glass substrate

Before loading the glass substrates into the deposition chamber (for Electron-beam ITO deposition), they were cleaned by the following steps:

a) Initially the glass substrate were dipped and vibrated in acetone by Ultrasonic Vibrator for 10 minutes to remove organic residues.
b) Then the substrates were cleaned with de-ionized (DI) water.
c) Again the substrate were dipped and sonicated in acetone by Ultrasonic Vibrator for 10 minutes.
d) After that the glass substrates were cleaned with de-ionized water.
e) And finally the substrates were dried in hot air.

2.3 Deposition of films by electron-beam evaporation

ITO thin films were deposited on a glass substrate at room temperature by electron beam evaporation technique using an Edwards E-306. The various quantities such as source to substrate height, deposition rate, beam current, chamber pressure, quality of substrate, substrate temperature, size of atomized particles etc. affect the film properties. Very thin layer of ITO (~ 100 nm) was deposited by electron-beam evaporation process with the chamber pressure of 2.5×10^{-5} mbar, beam current of 30 mA and voltage of 2 KV. After ITO deposition, the samples were annealed in a furnace (Carbolite CWF 12/13) in air at 600°C for 10 min [17]. In the same manner a layer of LaF_3 of about 5nm was deposited by electron-beam evaporation process with the chamber pressure of 2.5×10^{-5} mbar, beam current of 20 mA and voltage of 2 KV. Under this condition the deposition rate of the LaF_3 was found to 0.333 nms^{-1}. The aluminum layer of thickness about 50 μm was fabricated under the chamber pressure of 2.5×10^{-5} mbar, beam current of 20 mA and voltage of 4 KV.

2.4 Preparation of colloidal solution of Si-NCs with DCB

In traditional colloidal nanocrystal synthesis, the surfaces of nanocrystals are passivated with coordinating ligands to provide solubility of nanocrystals in the solvents. After nanocrystal-film formation, the ligands are usually removed or exchanged with shorter ligands to enhance the interaction between nanocrystals. The presence of ligands, though required for solubility of the NCs, is an impediment, since it requires additional processing steps. Removal of the ligands may also cause detrimental cracking of the NCs thin films, as the interparticle distance is reduced. However, in a previous study, we found that stable colloidal suspensions of Si-NCs could be produced under certain solution conditions without the use of ligands attached to the NCs surfaces [18]. Specifically, Si-NCs were found to form stable suspensions in 1,2-dichlorobenzene (DCB). For the formation of Si-NCs colloids, the following protocol was used: DCB was dried with molecular sieves and degassed by bubbling nitrogen for 60 min in a Schlenk line to reduce the oxygen and moisture level. Si-NCs were dispersed in DCB (ACROS) to form a cloudy, yet stable solution. The solution concentration was at 4 mg Si-NCs/ml DCB. To reduce Si-NCs agglomeration in the solution, the cloudy solution was sonicated for different time such as 30 min, 40 min, 50 min, and 60 min.

2.5 Device fabrication

A schematic of the layered structure of fabricated Schottky junction solar cells (with and without buffer layer) are shown in Fig. 1.

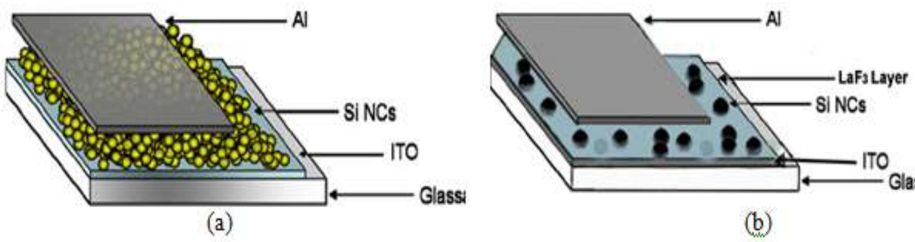

Figure 1: Schematic of the layered structure of schottky junction solar cells (a) without LaF_3 buffer layer, (b) with LaF_3 buffer layer

The fabrication of Schottky junction solar cells starts with an ITO film coated on a glass substrate. Traditionally ITO film has been used as a transparent conducting electrode in Schottky junction solar cell [5,19]. After the deposition of ITO layer the samples were annealed at 600°C for 10 min to reduce the surface roughness and to increase the transparency. After sonication in different time and amplitude, the colloidal solution of Si-NCs and DCB is applied on ITO coated glass by drop-casting method with micropipette. After drop-casting, the Si-NCs layer with DCB on ITO coated glass substrate was annealed at 80°C for 10 min to dry the layer. Then the samples were cooled down to room temperature. In the next step, a very thin layer of LaF_3 is deposited over the Si-NCs layer by E-beam evaporation technique (for the schottky junction solar cell with buffer layer). Again the whole structure (including LaF_3 buffer layer) was annealed at 400°C for 10 minutes to reduce the roughness of the fabricated LaF_3 layer. After that a layer of aluminum (thickness about 50 μm) was deposited as the cathode of the final device. Finally the samples were annealed in a thermal annealing furnace (carbolite CWF 12/13) in air at 150°C for 20 min to make the good connections at different layers of the fabricated device. After annealing they were left to be cooled naturally to the room temperature. Afterwards the whole structure was encapsulate with glass plate, which was glued on the top and brought out connection from anode and cathode by using copper wires (connected using silver paste).

2.6 Device characterization

Hitachi S-3400N scanning electron microscopy (SEM) imaging was used to study the surface morphology of the drop casted Si-NCs layers. The dependency of the absorption on layer thickness of the Si-NCs and photoluminance (PL) of the Si-NCs was studied using UV spectrophotometer of model SHIMADZU UV-1650PC. The C-V, G-V response were done by impedance analyzer. The elemental composition of the Si-NCs with and without LaF_3 was studied by EDX of model Inspect EDXA F50. Photo-current versus voltage (I-V) measurement was performed with a Keithley model 2400 source meter and a solar simulator system.

III. Results And Discussions

The dependency of the optical absorption on the particle size of the photoactive layer (Si-NCs) is important for the performance enhancement of the nanocrystals based Schottkey junction solar cells. The effect of Si-NCs size along with ITO layer on absorption was studied using U-V spectrophotometer with the wavelength range of 300-800nm. Fig. 2 shows the effect of absorption over wavelength for Si-NCs layer with different sonication time (which is correspond to the nano-crystals size). The absorption spectra of thin films obtained by drop-casting a colloidal solution of Si-NCs with DCB showed a significant change when the sonication time is changed. From this figure it is clear that the absorbance of the Si-NCs layer increases with increasing exility of the Si-NCs. In all cases the absorption is maximum above wavelength of 450nm.

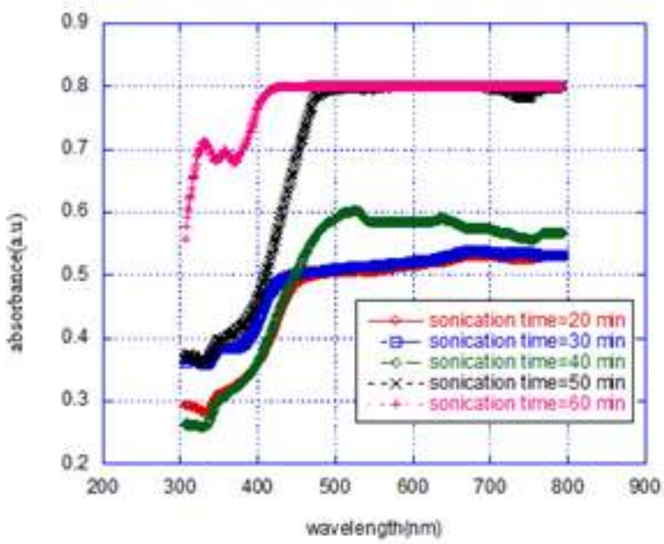

Figure 2: Absorption versus wavelength curve of Si-NCs layer at different sonication time.

The effect of the particles size of photoactive (Si-NCs) layer on photoluminace in the wavelength range of 200-800nm with excitation wavelength of 280nm was studied. Fig. 3 shows that effect. From the figure a PL peaking was found at the wavelength of about 570nm for all cases.

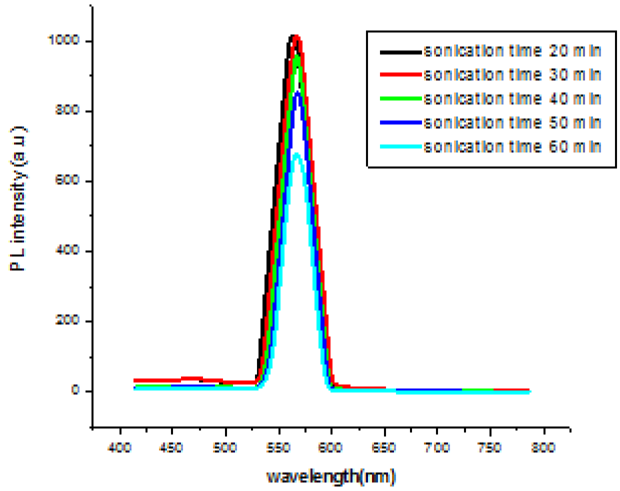

Figure 3: Effect of sonication time on the photoluminescence (PL) of the photo active layer.

As mention earlier the surface morphology of the Si-NCs deposited over the ITO coated glass was study by the Scanning Electron Microscope (SEM). Fig. 4 and Fig. 5 shows the SEM images of colloidal Si-NCs deposited sample on ITO coated glass at different sonication time (50 min and 60 min). From the above figure, the Si-NCs are clearly detected. The variation of Si-NCs size were investigated due to sonication time. From the figures it is observed that the average particles size for the sample of 60 min sonication time are smaller in comparison with samples of 50 min sonication time. It was also observed that the NCs sizes were in the diameter range from 45 nm - 130 nm depending on the sonication time.

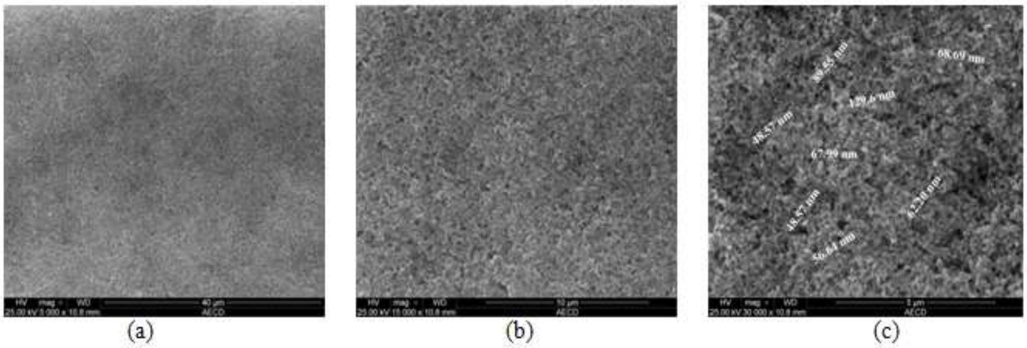

Figure 4: Surface morphology of Si-NCs layer without LaF_3 at sonication time 50 min with magnification (a) 5000 (b) 15000 and (c) 30000.

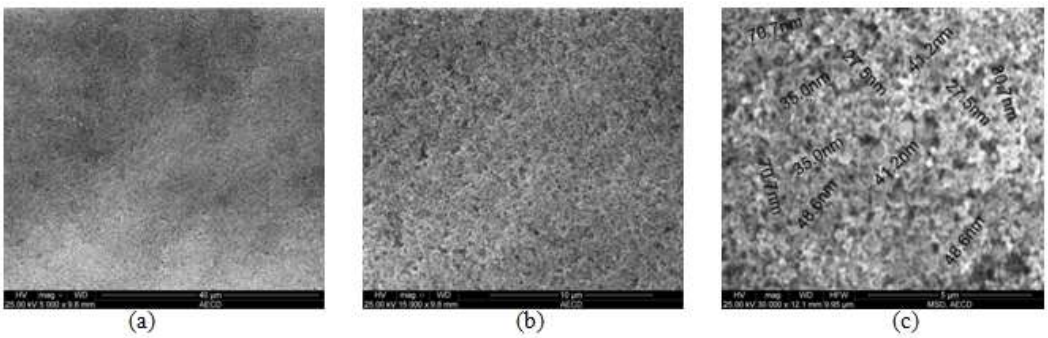

Figure 5: Surface morphology of Si-NCs layer without LaF_3 at sonication time 60 min with magnification (a) 5000 (b) 15000 and (c) 30000.

Fig. 6 shows the cross-sectional view of the fabricated device at 5000 times magnification that shows the various layers of the glass/ITO/Si-NCs hetero-structure. The ITO layer is not clearly visible as the thickness of ITO layer was around 200 nm whereas the thickness of the Si-NCs layer was about 35µm.

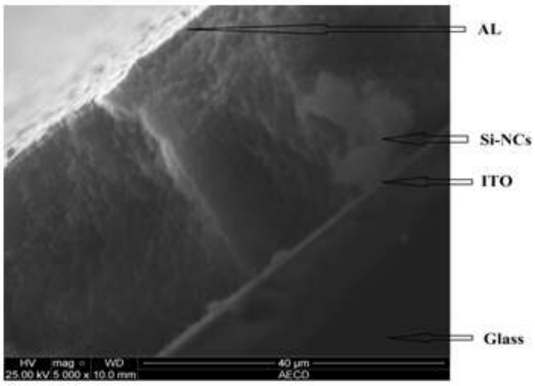

Figure 6: SEM image of cross-section for glass/ITO/Si-NCs/Al structure with 5000 magnifications.

EDX studies were used to analyze the elemental composition of the Si-NCs with and without LaF_3 deposited over ITO coated glass substrate. Fig. 7 shows the EDX of the sample with LaF_3 buffer layer. This confirmed the presence of Si-NCs embedded in non-stoichiometric LaF_3 layer. Fig. 8 indicates the strong presence of silicon with minor presence of some other compositional particles.

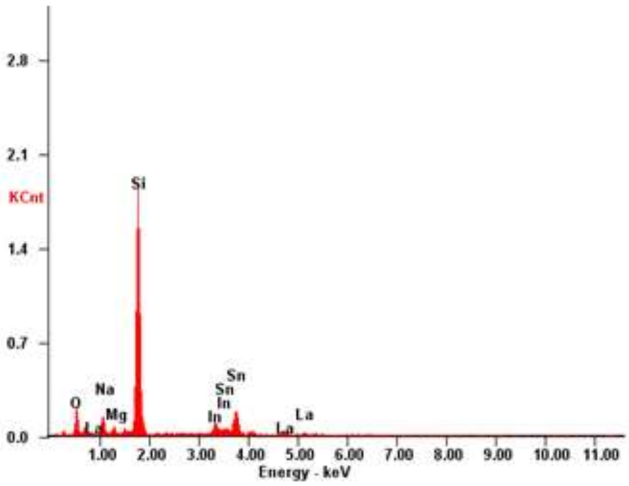

Figure 7: EDX spectra for Si-NCs layer embedded in LaF3 deposited on ITO coated glass.

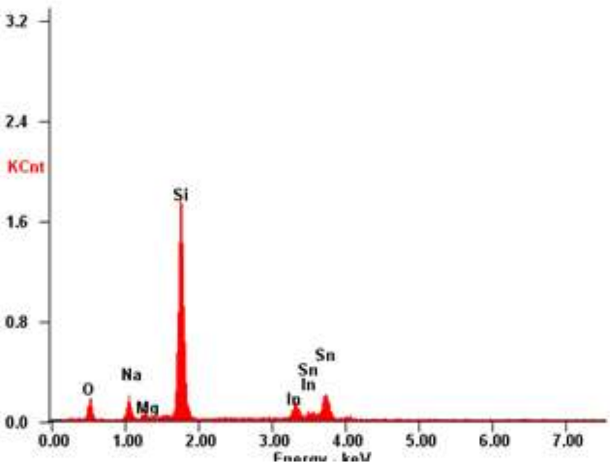

Figure 8: EDX spectra for Si-NCs layer deposited on ITO coated glass.

The I-V characteristics of the designed Al/Si-NCs/ITO without LaF_3 were studies under forward and reverse bias condition. The dark I-V characteristic of the fabricated schottky junction solar cell (Fig. 9) shows a typical rectifying junction behavior with threshold voltage of 0.3V. The reverse I-V characteristics under 1.5AM simulated light illumination were investigated without LaF_3 (Fig. 10) and with LaF_3 (Fig. 11) buffer layer to find the influence of the buffer layer on the performance of fabricated schottky junction photovoltaic cell. The I-V characteristic with LaF_3 buffer layer clearly indicates an enhancement (around 15 times compared to without LaF_3 structure) in the photo-generation and collection that is reflected in the enhancement of the reverse current of the device. Both I-V characteristics curve indicates poor short circuit current. This is because of the recombination of photo generated charges before they are reached at collectors.

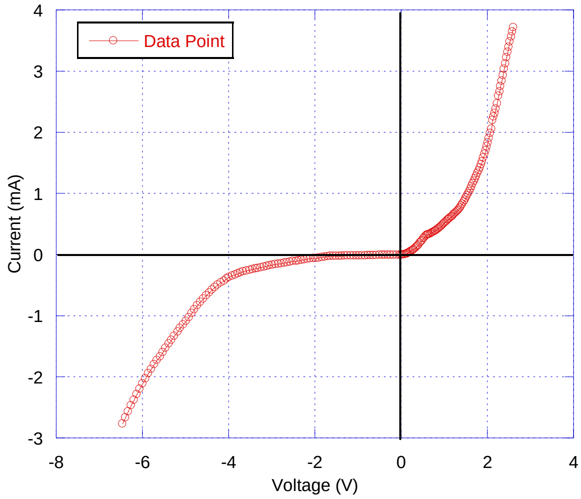

Figure 9: The I-V characteristics of the fabricated schottky junction solar cell under dark condition.

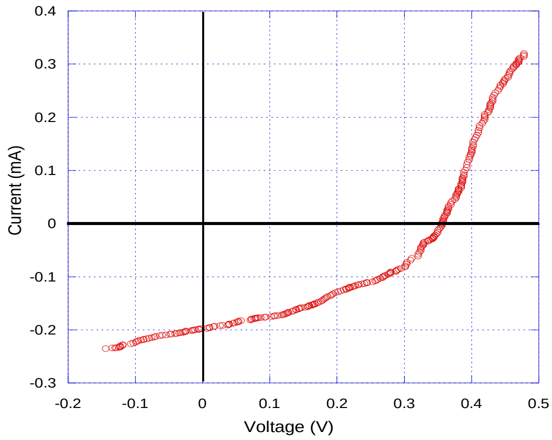

Figure 10: The I-V characteristics of the fabricated schottky junction solar cell under 1.5 AM light without LaF_3 buffer layer.

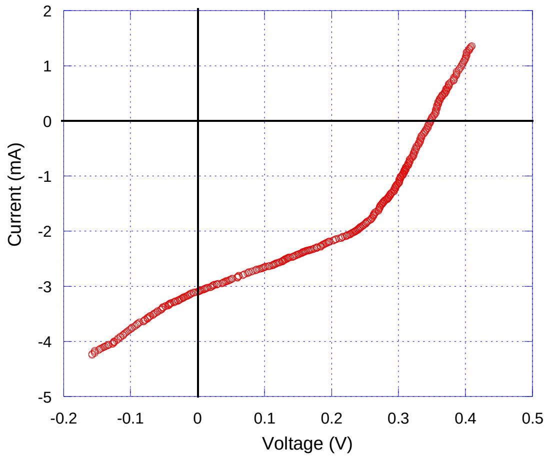

Figure 11: The I-V characteristics of the fabricated schottky junction solar cell under 1.5 AM light with LaF_3 buffer layer.

Fig. 12 shows the Capacitance-Voltage (C-V) curves of the Al/Si-NCs/ITO configuration. The C-V response indicates that the fabricated structure can acts as schottky junction device for various frequencies. C-V measurements were performed at room temperature using a standard C-V measurement setup. The C-V characteristics were measured over a bias voltage -4V to +4V and +4V to -4V. The DC voltage was applied to the top Al electrode with respect to the back ITO contact and superposed with various frequencies. In C-V curves clear peaks were observed in the inversion region, which were attributed to carrier exchange between the semiconductor substrates and bound states quantum confinement of carriers in the Si-NCs. The hysteresis loops were observed and the results indicate that the electrons were charged into Si-NCs.

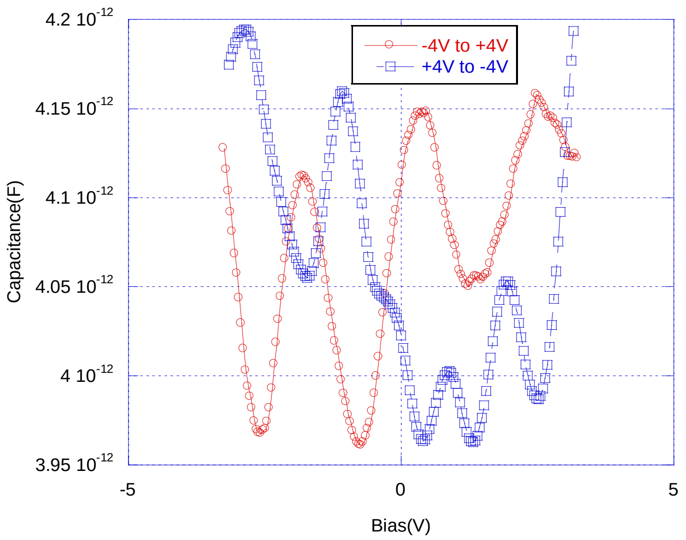

Figure 12: C-V hysteresis curves of Al/Si-NCs/ITO structure at frequency of 10 KHz.

Fig. 13 shows the G-V curve s of the Al/Si-NCs/ITO devices, measured at room temperature for various signal frequencies with bias voltage between -4V and +4V. The voltage was applied to the top Al electrode with respect to the back ITO. The fig. 13 clearly indicates that the conductance of the device is maximum for the frequency of 10 KHz.

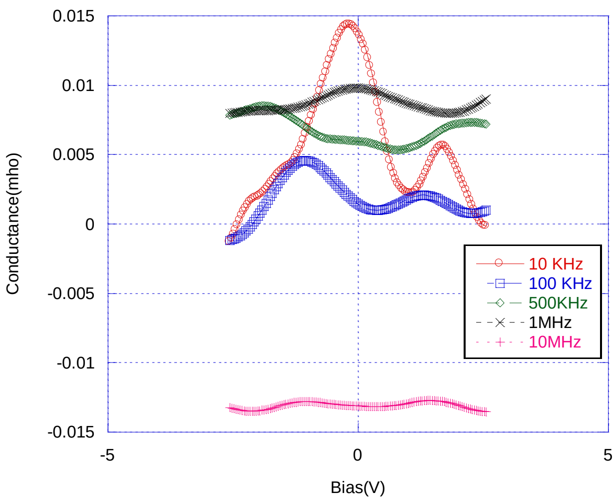

Figure 13: G-V hysteresis curves of Al/Si-NCs/ITO structure at different frequencies.

IV. Conclusions

Silicon Nano-Crystals can be fabricated at low temperature from solution processing without any vacuum equipment or high-temperature processing. Compared to Schottky junction solar cells reported previously, the whole fabrication method (in this work) for preparing Schottky junction photovoltaic is simple, cheap and fast. The performance parameters such as, JS_C, VO_C, FF and conversion efficiency was low for our fabricated cells. This was due to fabrication process used to fabricate the solar cell, processing environment, quality of the Si-NCs etc.

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References

[1]. Nathan S. Lewis, Powering the Planet, *MRS Bulletin, Volume 32,* pp 808-820, October 2007.
[2]. Luque A., Hegedus S., *Handbook of Photovoltaic Science and Engineering*, Wiley, Chichester; 2003.
[3]. Chang J, Sun B, Breiby DW, Nielsen MM, Sölling TI, Giles M, McCulloch I, Sirringhaus H, Enhanced mobility of poly (3-hexylthiophene) transistors by spin-coating from high-boiling-point solvents, *Chemistry of Materials, Volume 16,* pp 4772-4776, November 2004.
[4]. H. Sirringhaus1, P. J. Brown, R. H. Friend, M. M. Nielsen, K. Bechgaard, B. M. W. Langeveld-Voss, A. J. H. Spiering, R. A. J. Janssen, E. W. Meijer, P. Herwig & D. M. de Leeuw, Two-dimensional charge transport in self-organized, high-mobility conjugated polymers, *Nature, Volume 401,* pp 685-688, October 1999.
[5]. Luther, J. M., Law, M., Beard, M. C., Song, Q., Reese, M. O., Ellingson, R. J., Nozik, A. J., Schottky Solar Cells Based on Colloidal Nanocrystal Films, *Nano Letters, Volume 8,* pp 3488-3492, 2008.
[6]. Keith W. Johnston, Andras G. Pattantyus-Abraham, Jason P. Clifford, Stefan H. Myrskog, Dean D. MacNeil, Larissa Levina, and Edward H. Sargent, Schottky-quantum dot photovoltaics for efficient infrared power conversion, *Applied Physics Letters, Volume 92,* pp 151115, April 2008.
[7]. T. Shimizu-Iwayama, K. Fujita, S. Nakao, K. Saitoh, T. Fujita, and N. Itoh, Visible photoluminescence in Si+ -implanted thermal oxide films on crystalline Si, *Journal of Applied Physics, Volume 75,* pp 7779-7783, 1994.
[8]. K. S. Min, K. V. Shcheglov, C. M. Yang, Harry A. Atwater, M. L. Brongersma and A. Polman, Defect-related versus excitonic visible light emission from ion beam synthesized Si nanocrystals in SiO_2, *Applied Physics Letters, Volume 69,* pp 2033, 1996.
[9]. K. A. Littau, P. J. Szajowski, A. J. Muller, A. R. Kortan, and L. E. Brus, A luminescent silicon nanocrystal colloid via a high-temperature aerosol reaction, *Journal of Physical Chemistry, Volume 97,* pp 1224-1230, 1993.
[10]. D. M. Holunga, R. C. Flagan, and H. A. Atwater, A Scalable Turbulent Mixing Aerosol Reactor for Oxide-Coated Silicon Nanoparticles, *Industrial & Engineering Chemistry Research, Volume 44,* pp 6332-6341, 2005.
[11]. Q. Zhang, S. C. Bayliss, and D. A. Hutt, Blue Photoluminescence and Local-Structure of Silicon Nanostructures Embedded in Sio_2 Matrices, *Applied Physics Letters, Volume 66,* pp 1977-1979, 1995.
[12]. J. U. Schmidt and B. Schmidt, Investigation of Si nanocluster formation in sputter-deposited silicon sub-oxides for nanocluster memory structures, *Materials Science and Engineering, B, Volume 101,* pp 28-33, 2003.
[13]. L. A. Nesbit, Annealing characteristics of Si-rich SiO_2 films, *Applied Physics Letters, Volume 46,* pp 38-40, 1985.
[14]. F. lacona, G. Franzo and C. Spinella, Correlation between luminescence and structural properties of Si nanocrystals, *Journal of Applied Physics, Volume 87,* pp 1295-1303, 2000.
[15]. U. Kahler and H. Hofmeister, Visible light emission from Si nanocrystalline composites via reactive evaporation of SiO, *Optical Materials, Volume 17,* pp 83-86, 2001.
[16]. A.T.M. Saiful Islam and Abu Bakar Md. Ismail, Fabrication of uniform-macroporous silicon and its possible application in hybrid solar cell, *International Conference on Advances in Electrical Engineering (ICAEE), pp 401-404,* December-2013, Dhaka, Bangladesh.
[17]. A.T.M. Saiful Islam, Mushtaq Ahmed Sobhan and Abu Bakar Md. Ismail, The effect of introducing Macroporous Silicon as the Cathode of Bulk Heterojunction Hybrid Solar Cell, *International Conference on Materials, Electronics & Information Engineering (ICMEIE),* June-2015, Rajshahi, Bangladesh.
[18]. Liu CY, Holman ZC, Kortshagen U., Hybrid solar cells from P3HT and silicon nanocrystals, *Nano Lett., Volume 9,* pp 449-452, 2009.
[19]. J. Tang, X. Wang, L. Brzozowski, D. A. R. Barkhouse, R. Debnath, L. Levina and E. H. Sargent, Schottky quantum dot solar cells stable in air under solar illumination, *Advanced Materials, volume 22,* pp. 1398 - 1402, 2010.

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