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Fabrication of Non-volatile Charge Storage Memory Device by Novel doped ZnO nanoparticles with 4.79 eV bandgap

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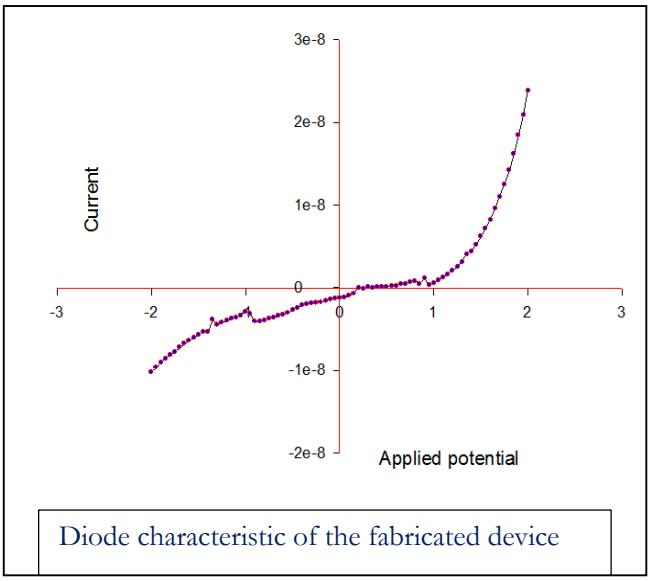
Abstract

Nowadays, the drastic participation of the nanosized materials in technology have been implicated with various applications which mainly aims the performance optimization, dimensional down scaling, ultra-low power consumption etc. to overcome the fundamental limits of the microscale devices. It is the requisite stage to familiarize a convenient alternative of the traditional large scale technologies in purpose of accelerating the flow of the applied science for mankind. This article presents two novel non-volatile device structures which are fabricated by layer by layer deposition method. I-V measurements for both the samples justify the device characteristic as p-type – insulator–n-type configuration. The measurements confirm that the successful fabrication of those devices and proves the high density charge capacity with improved lifetime of the carriers compared to erstwhile reports. The measured the threshold voltage for this device is 0.939V.



Diode characteristic of the fabricated device

Keys: Zinc oxide, charge storage devices, quantum physics, nanotechnology

SOUDIP SINHA ROY

1 Introduction

To unfold the hidden mysteries of the molecules and their applications the nanoscience have been presented plenty of its morphism in order to rapid performance scale up with drastic dimensional down scaling. Manipulation of the molecular information technology has numerous existing approaches for instance molecular non-volatile memory, electron spin devices, nano-memorister, quantum dot island based devices, capacitive devices etc. which have proven their superior activities in binary logic and storage device applications. Nowadays, the utility of the hard storage has become quite crucial and instantaneous which claims to be tinier and fastest to meet the present requirements of the mankind. The fabrication of the non-volatile charge storage devices opens a convenient way with millions charge storing capacity in a few nanometer area.

In this letter the ZnO nanoparticle based non-volatile memory device has reported which provides a better optimization to the device efficiency. To fabricate this device three major materials have involved those are PMMA, Ag NPs, ZnO NPs. In another type of the device is fabricated by using nafion which exhibits a benchmarking of the p-i-n diode. The PMMA layer is used as the tunneling barrier for the electrons that provides a path to the electrons to be tunneled during the biasing application but resistance during cutoff state. It also acts as the high impedance to the electrons during cutoff state that resists from the discharging by defending the reverse tunneling. To fabricate this device the ZnO and Ag doped ZnO NPs are utilized and specifically sputtered on the surface of the ITO. Two different types of devices have been walked through firstly ITO/PMMA/Nafion has been examined and next is ITO/PMMA /ZnO-NPs doped with Ag NPs. The reduced bandgap of the doped ZnO layer works as the electron hopping state and allows to operate it at ultra-low power input.

2 Background of Non-volatile Memory Devices

Since the last twenty years of the 19th century the advancement of the nanotechnology has bequeathed a lot of novel applications to the current technology to accelerate its current flow over its fundamental limits. The molecular technology has eliminated various complications in the device fabrication and operation for instance impurity inconsistency, thermal carrier diffusion, elevated outlay of photolithography etc. which genuinely overcomes the unwanted faults in circuit and strictly shrinks the number of superposition states. In the recently published article it is reported that [1,2] the tunneling current depends on the tunneling barrier resistivity which get violated dynamically due to the operating temperature variation. Therefore, the temperature stability is also a key point which rights to be high to opt the ultimate device performance.

The synthesis of the non-volatile memory devices through the molecular thin film technology has already been taken care of before a couple of years. The floating gate MOS devices also exhibits acceptable performances in charge storing but those have the less storage density and lower speed of operation. The involvement of the nanoparticles induce a high surface area with the wide bandgap that imposes to store high density data and optimized operating speed with ultralow power consumption and minute faults.

ITO/PMMA/Ag NPs based device has already been reported previously which exhibits superior performance adaptability [2]. The ON/OFF ratio (switching rate) has also improved drastically due to the incorporation of the PMMA layer in between the ITO and the Ag NPs layer. Another report communicates that the structure ITO/a-C/ZnO/a-C/Al contact also exhibits very high optimization in switching rate and the charge storage capability where two a-C has incorporated which acts as a typical insulator [1].

3 Preparation of respective nanoparticles

The following nanoparticles are used in the device fabrication process. The process of synthesis of those nanomaterial are very easy, cheaper and time efficient. The involved chemicals have easy commercial availability. The list of chemicals are as follows.

Sodium Borohydrate (NaBH_4); AgNO_3; PVP; Zinc Acetate-dihydrate (Zn$(COOCH_3)_2.2H_2O$); Isopropyl Alcohol ($CH_3CH_2CH_2OH$); Ethanolamine (C_2H_7NO).

3.1. ZnO NP preperation method

As a precursor commercially available Zinc Acetate-dihydrate (Zn$(COOCH_3)_2.2H_2O$) salt was used and dissolved into Isopropyl Alcohol ($CH_3CH_2CH_2OH$) having molar mass 60.09 g/mol. The final obtained solution was 0.5M. Afterwards, the solution was put in stirring and the Ethanolamine (C_2H_7NO) was added drop-wise during stirring at constant 80 °C temperature until the gel is formed. After obtaining the gel the sample was dried at 35°C until the solvent is completely evaporated out. However, the organic solvent evaporates faster in rate even at room temperature.

3.2. Ag NP preperation method

With the 60ml of distill water the 0.004M NaBH_4 was dissolved and kept for 30 min at continuous stirring in an ice bath. During stirring gently add 4ml of 0.002M AgNO_3 dropwise until the color of the solution is converted into light yellow. Once the desired color is achieved 0.3% PVP as the capping agent was gently added. Afterwards, store the prepared solution immediately in dark space to avoid the particle agglomeration.

3.3. Ag doped ZnO nanopowder preperation method

As a precursor commercially available Zinc Acetate-dihydrate (Zn$(COOCH_3)_2.2H_2O$) salt was used and dissolved into Isopropyl Alcohol ($CH_3CH_2CH_2OH$) having molar mass 60.09 g/mol. The final obtained solution was 0.5M. Afterwards, the solution was put in stirring and the Ethanolamine (C_2H_7NO) was added drop-wise during stirring at constant 80 °C temperature until the gel is formed. After the ZnO gel formation the 5% concentric AgNO_3 was added drop wise while stirring at 6000 rpm inside an ice bath. Once the doping is successful then by centrifuging the sample and washing by MEA several times bright white colored Ag doped ZnO nanopowder was obtained.

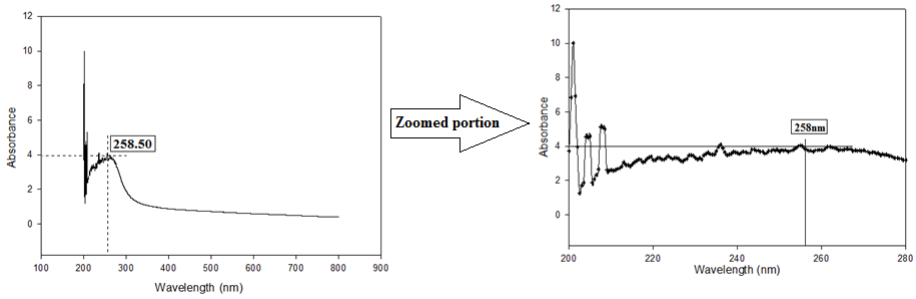


Figure 1 | (a) UV-vis measurement for the pure ZnO NPs (50nm), (b) Zoomed view for further clarification

As the UV visible optical bandgap approximation asserts that B.G.= $\frac{1240}{\lambda}$ eV.

Therefore, the calculate B.G. of this above ZnO sample is given by 1240/258.40= 4.79 eV.

The bandgap for a pure ZnO nanoparticle is at around ~3.2eV which responds to 380nm UV wavelength. But in this case the bandgap is found with an unexpected increment which responded at 258.40 nm UV wavelength**.** *In the next context this reason will be revealed out. Research is going on, on this topic.*

3 Device Fabrication

The fabrication is the ultimate step for providing a physical aspect to any theoretical substance. In this case two different types of devices have been fabricated fig. 2 is the ITO/PMMA/ZnO NPs based device. This device operates with the basic fundamental process of the charge trapping and hopping states. But the second device fig. 3 operates with the reduced bandgap of the ZnO through Ag doping and results the ultra-low power operating with optimized ON/OFF ratio.

4.1 ITO/PMMA/Naffion structured device

Chemical formula of the nafion is $C_7HF_{13}O_5S{\cdot}C_2F_4$. Initially the ITO was repeatedly cleaned by the concentric ethanol and the acetone in the ultrasonicator then it was dried at and hour at room temperature. After that the ITO was exposed to the PMMA by a suitable spin coater and kept for 24 hrs. at room temperature for drying purpose. The grown thickness in 40nm. Once the drying is completed then highly purified nafion was drop casted onto the dried PMMA surface of the ITO and dried for 24 hrs. at room temperature.

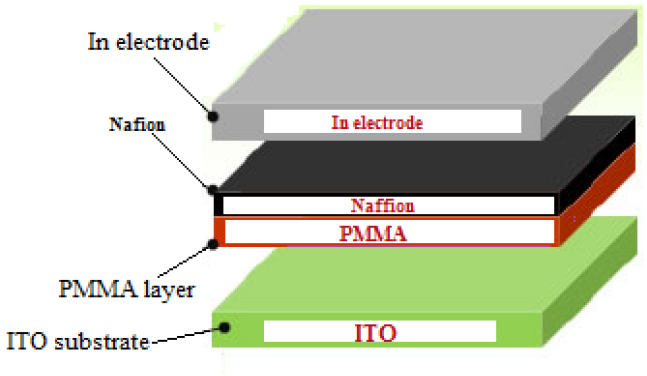


Figure 2 | ITO/PMMA/Nafion structured device

4.2 ITO/PMMA/Ag doped ZnO structed device

The fabrication process

Initially the ITO was repeatedly cleaned by the concentric ethanol and the acetone in the ultrasonicator then it was dried at and hour at room temperature. After that the ITO was exposed to the PMMA and kept for 24 hrs. at room temp. for drying purpose. Once the drying is completed then the pre-synthesized Ag doped ZnO nanopowder was diluted into the 2 propanol and was sputterd onto the dried PMMA surface of the ITO and 100 nm thicker thin film was produced. After that the sample was kept at vacuum for 48 hrs in special purpse.

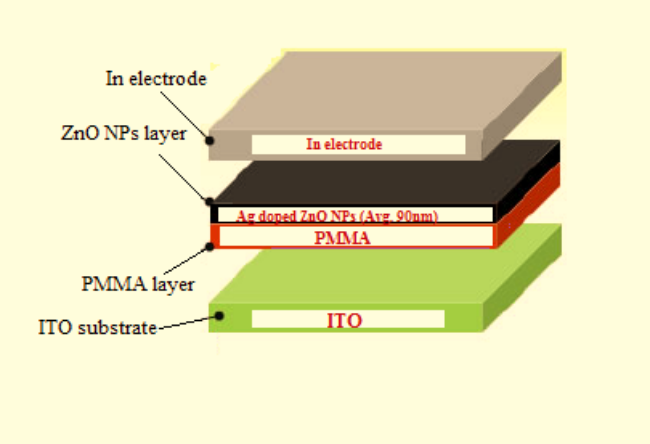


Figure 3 | ITO/PMMA/Ag NPs/Ag doped ZnO structured device

5 Performance Justification Through I-V and C-V Measurement

5.1 I-V measurement of the naffion based device.

I-V measurement is the technique which allows to investigate the current verses voltage mapping for any multipolar device. In this case both the fabricated devices are the bipolar systems those are verified through I-V measurements.

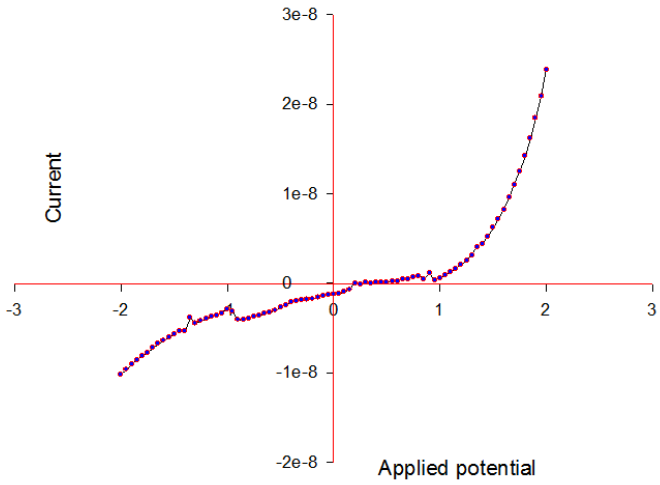


Figure 4| I-V measurement curve for the ITO/PMMA/Nafion based device

From above graph fig. 4, it is well visible that the curve is similar to the semiconductor diode where the electrons forward current is 2.39889e-9 A. The current is high compared to the previously reported devices [2]. This high current stands for the high probability of the electrons to be found in the conduction band of the nafion. Once the electrons are subjected to the electro motive field then gradually those try to overcome the PMMA barrier interface (thickness 250nm) and after a certain voltage the tunneling phenomena of the electrons are observed which is clear from the fig. 3. Within the potential range of 0.831V to 0.939V there has a transition of the electron which gives a small valley peak that satisfies the electron tunneling effect through the thin PMMA film. The upper valley provides 1.422e-9 A current which instantly falls down at 1.4444e-10 A. From 0.939V this device exhibits the standard diode characteristics. Therefore, the threshold voltage for this device is 0.939V. The measurement form -2V to +2V assures that this diode characteristic is well justified and meet with the standard semiconductor devices.

5.2 C-V measurement of the Nafion based device.

Cyclic voltammetry measurement for the nafion based sample. Keeping K+ ions in the solution the CV measurement was performed.

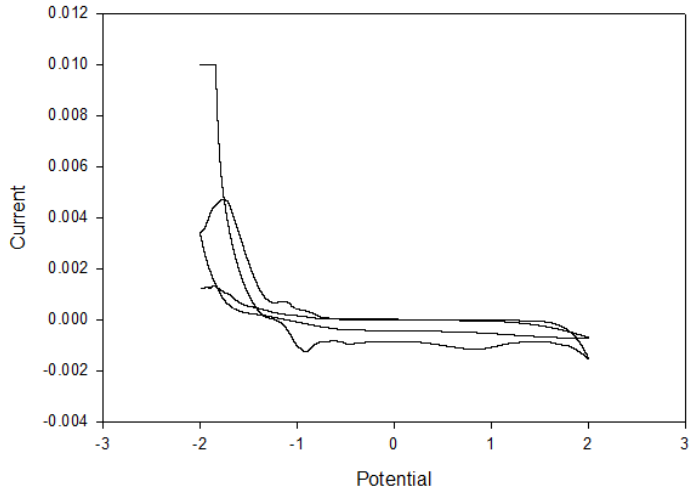


Fig. 5: I-V measurement curve for the ITO/PMMA/Nafion based device with the I-V four probe measurement system.

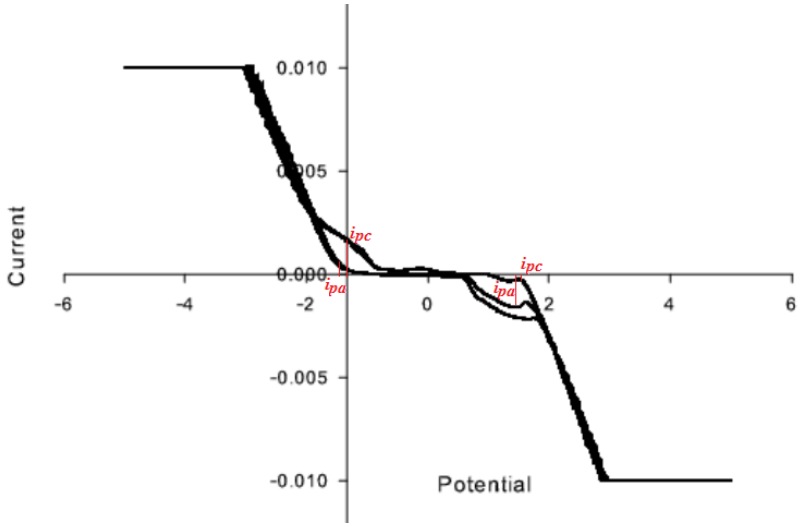


Figure 6| C-V measurement curve for the ITO/PMMA/Nafion based device

According to the C-V analysis if the device is fully reversible then obviously ipc/ipc should be equal to 1. But in this case this ration deviates from one by the factor of 4.3170. The anodic current ipa is 0.000351429A and the cathodic current is 0.00151714A. *In case of forward cycle the current is comparative high to the reverse cycle. Therefore, it is identified that this device is having a non-volatility nature.* This non-volatile nature is in form of charge shoring capacity. The specified interval between two cycles forward and reverse is 2sec.

5.3 I-V measurement of the Ag doped ZnO based device.

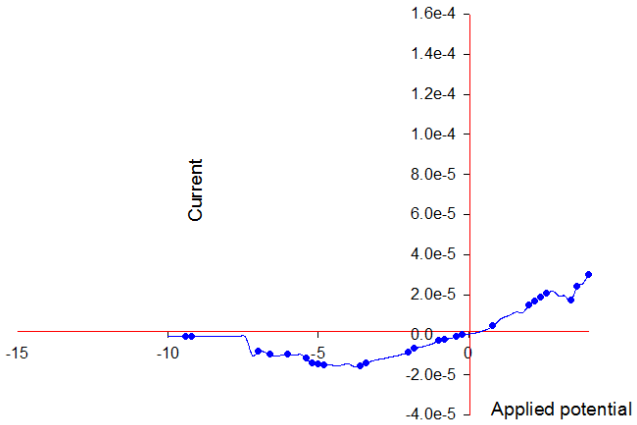


Figure 7 | I-V measurement curve for the ITO/PMMA/Ag doped ZnO NPs based device.

From this measurement curve figure 7, it is seen that that curve is following the standard p-i-n characteristics. The incorporation of the intermediate insulating layer differs the device from the general p-n diode semiconductors where the p-type and the n-type semiconductors are attached and separated by the self-induced insulating layer called depletion layer. But when this depletion layer is mask fabricated then the device characteristics changes to the p-i-n characteristics where the electron tunneling takes place through the insulating layer. Depending on the thickness of the tunneling barrier tunneling current varies and gives a search tunneling current which typically provides an oscillation as shown in the figure 8. According to the measurement curve the maximum stable current output is obtained at 3.984V is 3.2e-5 A. The measured threshold voltage for this device is 0.339V.
The full frame of the graph is shown below, where the highlighted part of the graph is having a noise like phenomena which is mostly caused by the unwanted and uncontrolled electron tunneling within the applied potential range, figure 8.

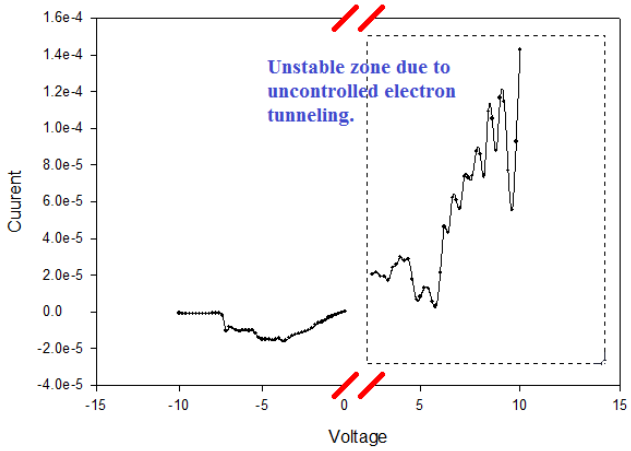


Figure 8 | Complete curve of I-V measurement curve for the ITO/PMMA/Ag doped ZnO NPs based device.

6. Conclusion

This work is performed for fabricating the non-volatile devices based on ZnO and metal doped ZnO nanoparticels. In collaboration of the four probe I-V characteristics measurement the devices have proven that both are having an non-volatile nature and the those are having very less threshold voltage which can trigger device at a very low power. The improvent in the charge storage capacity is notable in both the devices. The unexpected bandgap for the ZnO is being verified through several observations and experiments. In the future articles this may come across.

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