



Master's Thesis 석사 학위논문

Metal-Insulator-Metal tunnel diode with various materials and structural designs for rectifying effect

Su Jin Heo(허 수 진 許 秀 珍)

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Advisor: Professor Jae Eun Jang Co-advisor: Professor Chang Hee Cho

By

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A thesis submitted to the faculty of DGIST in partial fulfillment of the requirements for the degree of Master of Science in the Department of Information and Communication Engineering. The study was conducted in accordance with Code of Research Ethics¹

12.21.2018

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Metal-Insulator-Metal tunnel diode with various materials and structural designs for rectifying effect

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Accepted in partial fulfillment of the requirements for the degree of Master of Science.

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ABSTRACT

Electrical characteristics of metal-insulator-metal (MIM) tunnel diodes with various materials and structural designs for high speed operation and rectification effects have been studied. Recently, high speed devices have been demanded in many fields such as high performance control process unit (CPU), optoelectric devices and communications. Among various electrical components for high speed working, in here, rectifying diodes have been studied as basic building block structure for the high-speed devices.

Traditionally, PN junction diodes and Schottky diodes have been studied intensively for rectifying function. However, the mechanism of the pn junction diode is not suitable for high speed rectification due to the large depletion region width. For Schottky barrier diode, it is more suitable for high-speed switching. Since it has very short reverse recovery time. However, the driving frequency limit of Schottky diode is around 5THz. Therefore, new rectifying mechanism operating at a high frequency should be suggested. As one of the rectifying diode, MIM diode is operated by quantum electron tunneling effect which occurs when a particle passes through a thin potential barrier. So, MIM tunnel diode is enabled high-speed switching and it can have a low operating voltage.

In order to increase the high-speed operation and rectification effect of MIM diode structure, the MIM diode should have a thin insulator layer as a tunneling barrier. Moreover, the current-voltage characteristics of the devices should have non-linearity and asymmetry. However, due to the tunneling mechanism of device structure, the non-linearity or asymmetry characteristics should be improved. Therefore, this paper was focused to improve the non-linearity and asymmetry characteristics of MIM diode. To improve the non-linearity and asymmetry characteristics of the diode, the various MIM diode structure were studied different work function of metal, double insulator layers and device design as important parameters. The simple flat MIM didoes, metal-insulator-insulator-metal (MIIM) diode, and vertical metal cylinder structure (VMCS) MIM diode have been studied. The simple flat MIM diode considering material effect together did not show high non-linearity and asymmetric characteristics. However, the flat MIIM diode has good non-linearity and some asymmetry in there I-V characteristics. Although VMCS MIM diode did not have a better non-linearity and asymmetry properties. However, the VMCS MIM diode shows the non-linearity and asymmetry characteristic more improved than simple flat MIM tunnel diode. Therefore, we could show the feasibility of high speed switching and rectification in MIM diode using the vertical metal cylinder structure which is applied the device design as a parameter. These new MIM diode structure can be applied to various field such as new electronic devices, rectifier antennas for energy harvesting, optical devices, infrared/terahertz detectors in high frequency range, and electric high-speed switching devices.

Keywords: MIM diode, MIIM diode, Vertical Metal Cylinder Structure, non-linearity, asymmetry

List of Contents

Abstract ·····i
List of contents ·······ii
List of tables ·······iii
List of figures ······ vi

I . INTRODUCTION

1.1 Theoretical background1
1.1.1 PN junction diode & Schottky barrier diode1
1.1.2 Tunnel diode
1.1.2.1 Quantum tunneling effect
1.1.2.2 Tunnel diode (Esaki diode)
1.1.3.MIM tunnel diode
1.1.3.1 Conduction mechanism of MIM diode
1.1.3.2 Characteristics of MIM diode10
1.1.3.3 Theoretical model of MIM diode12
1.1.3.4 Structural tendency of MIM diode14
1.2 Objective
II. FABRICATION
2.1 Fabrication of simple flat MIM diode17
2.2 Fabrication of flat MIIM diode21
2.3 Fabrication of vertical metal cylinder structure MIM diode24
III. ELECTRIC CHARACTERISTICS AND RESULT
3.1 Electrical characteristics of simple flat MIM diode27
3.2 Electrical characteristics of flat MIIM diode
3.3 Electrical characteristics of vertical metal cylinder structure MIM diode40
IV. CONCLUSION
REFERENCE

List of tables

Table 2.1 The samples of simple flat MIM diodes 18
Table 2.2 The samples of flat MIIM diodes 22
Table 3.1 Work function of metal materials 27
Table 3.2 The properties of oxide materials 27
Table 3.3 Real hole diameter of the cylinder structure

List of figures

Figure 1.1 A pn junction diagram at zero-bias1
Figure 1.2 Schematic and band diagram of schottky diode2
Figure 1.3 Quantum tunneling phenomenon through the potential barrier
Figure 1.4 Electrical characteristic of Esaki tunnel diode
Figure 1.5 Energy diagram of Esaki diode at each point in I-V characteristic
Figure 1.6 SEM image of Cat-whisker Point contact diode7
Figure 1.7 Energy band diagram of asymmetrical metal MIM diode at zero bias, forward
bias, and reverse bias
Figure 1.8 Energy band diagram of the direct tunneling and the Fowler-Nordheim
Tunneling
Figure 1.9 Theoretical tunnel resistance as the applied bias for an asymmetrical MIM
Structure
Figure 1.10 Equivalent circuit of MIM diode12
Figure 1.11 The schematic and SEM image of the edge MOM diode14
Figure 1.12 The schematic and SEM image of lateral MIM diode15
Figure 1.13 The schematic and SEM image of later MIM diode15
Figure 1.14 The schematic of bent metal wire MIM diode15
Figure 2.1 The schematic of the simple flat MIM diode17
Figure 2.2 The schematic of the fabrication process of the simple flat MIM diode19
Figure 2.3 The schematic of the flat MIM diode
Figure 2.4 The schematic of the fabrication process of the flat MIIM diode23
Figure 2.5 The whole devices pattern design for vertical metal cylinder structure MIM
diode25
Figure 2.6 The schematic of fabrication process of the VMCS MIM diode26
Figure 3.1 Optical microscope image of simple flat MIM diode27
Figure 3.2 Energy band diagram of the simple MIM diodes at zero bias
Figure 3.3 I-V curves of the Al-Al_2O_3-Al diode $\hfill \sim 29$
Figure 3.4 Energy band diagrams of the Al-Al ₂ O ₃ -Al and Al-Al ₂ O ₃ -Au diodes at positive
bias
Figure 3.5 I-V curves of the Al-Al_2O_3-Au diode by the cross area $\cdots \cdots 30$
Figure 3.6 I-V curves of the comparing with Al-Al_2O_3-Al and Al-Al_2O_3-Au diodes $\ \cdots \cdots 32$
Figure 3.7 Asymmetry plots of the comparing with Al-Al $_2O_3$ -Al and Al-Al $_2O_3$ -Au diodes 32
Figure 3.8 FN tunneling model of Al-Al_2O_3-Al and Al-Al_2O_3-Au diodes at positive bias $\cdot33$
Figure 3.9 Energy band diagram of simple flat MIIM diode at zero bias
Figure 3.10 Energy band diagram of simple flat MIIM diode, Al-Al ₂ O ₃ -HfO ₂ -Al34
Figure 3.11 Energy band diagram of simple flat MIIM diode, Al-Al ₂ O ₃ -HfO ₂ -Au34

$Figure \ 3.12 \ I-V \ curves \ of \ the \ Al-Al_2O_3-HfO_2-Al \ and \ Al-Al_2O_3-HfO_2-Au \ diodes \ \cdots \cdots 36$
Figure 3.13 I-V curves of the comparing with Al-Al ₂ O ₃ -HfO ₂ -Al and Al-Al ₂ O ₃ -HfO ₂ -Au
diodes
Figure 3.14 Asymmetry plots of the comparing with Al-Al ₂ O ₃ -HfO ₂ -Al and Al-Al ₂ O ₃ -
HfO ₂ -Au diodes
Figure 3.15 FN tunneling model of Al-Al ₂ O ₃ -HfO ₂ -Al and Al-Al ₂ O ₃ -HfO ₂ -Au diodes at
positive bias
Figure 3.16 Schematic of flat MIM diode design for RF measurement
Figure 3.17 The schematic of VMCS MIM diode40
Figure 3.18 Optical microscope and SEM image of the VMCS diode and cylinder array $\cdot 41$
Figure 3.19 J-V curve of VMCS MIM diode with Al metal (Inset is I-V plot)41
Figure 3.20 SEM images of Al metal cylinder at the top view
Figure 3.21 SEM images of Au metal cylinder height and diameter43
Figure 3.22 J-V curve of VMCS MIM diode with Au metal cylinder44
Figure 3.23 Comparison the asymmetry of the VMCS diode with flat MIM diode45
Figure 3.24 FN tunneling model of VMCS MIM diode with Au metal cylinder at positive
Bias (Inset is FN characteristic at negative bias)45
Figure 3.25 SEM images of the vertical metal cylinder array with different pitch46
Figure 3.26 J-V characteristic of VMCS MIM diodes with different cylinder array pitch $\cdot\cdot46$
Figure 3.27 SEM images of 50 nm hole diameter cylinder array47
Figure 3.28 J-V characteristic of 4 aspect ratio cylinder MIM diode47
Figure 3.29 Asymmetry of the VMCS diode with 4 aspect ratio cylinder48

I. INTRODUCTION

1.1 Theoretical background

1.1.1 PN junction diode & Schottky barrier diode



Figure 1.1 A pn junction diode diagram at zero-bias

PN junctions are commonly used in diode, transistor and solid state electronics. When ptype and n-type semiconductor are junction, holes in p-region and electrons in n-region are recombination on the middle of the junction. So in the middle, the neutral equilibrium state is break down. There are depletion region or space charge region that are only remained the space charges exclude the carriers (holes and electrons). The junction is shown the rectification characteristics from the depletion region. Rectification is blocking the current at reverse bias and flow the current across the depletion region at forward bias. PN junction diodes are usually used as a convert alternating current (AC) to direct current (DC). However, the width of depletion region is large, so this mechanism is not suitable the high speed rectification. [1, 2]



Figure 1.2 Schematic and band diagram of schottky diode [3]

Schottky barrier diode is a junction of metal and semiconductor. Because the schottky diode does not have accumulation effect the current flows through the majority carriers rather than the minority carriers. So the schottky diode has very short reverse recovery time. Therefore, it is suitable for high-speed switching. Other advantages of the schottky diode are lower forward resistance and low noise generation. [4] For this reasons, the forward current has low operating voltage. In addition, schottky diodes are suitable for microwave receiver detectors and mixers. Because they generate lower noise. However, the driving frequency of schottky diodes is limited to 5THz. Therefore, a new approach to use at higher frequencies is needed. [5]

1.1.2 Tunnel diode

1.1.2.1 Quantum tunneling effect



Figure 1.3 Quantum tunneling phenomenon through the potential barrier [6]

The quantum-mechanical tunneling occurs when a particle passes through a thin potential barrier without overcoming it without any energy change. [5] This barrier is assumed to be higher than the kinetic energy of the particles and cannot be explained based on the law of classical mechanics. [7] According to classical physics, if the energy of the incident electron is less than the potential barrier, the electron is reflected. However, according to the quantum mechanism, the electron wave function has a finite probability of penetrating if the thickness of the potential barrier extremely thin. This phenomenon of electron penetration is called the tunnel effect. [5] Also, due to this tunneling effect, the probability that electrons are on opposite sides of the potential barrier is not zero. [8] This tunneling is characterized by a current flow across a thin semiconductor p-n junction (Esaki tunneling), a current flow through a thin oxide film (Metal-Insulator (Oxide)-Metal tunneling), electrical breakdown in solid dielectric (Zener tunneling). [5]

1.1.2.2 Tunnel diode (Esaki diode)

The Esaki tunnel diode was invented in 1957 by Japanese physicist Esaki Leona. This is a p-n junction diode applying the phenomenon that the negative resistance characteristic in the impurity semiconductor. The negative resistance is the phenomenon when the voltage increases but the current decreases. When the impurity concentration of the p-n junction is increased, the barrier of the depletion layer becomes very thin. Then tunneling effect occurs, the current flows suddenly and the negative resistance characteristic appears in the forward bias state. At this time, the doping concentration is mainly 10^{19} /cm³ and the depletion region must be very thin. [9, 10]

Figure 1.4 shows I-V characteristics of tunnel diode focused on forward bias. Based on the current axis, the tunneling determined the position between the origin and the valley point. The tunneling current increases to the valley point and after the valley point the current flows by overcoming the barrier. [2]



Figure 1.4 Electrical characteristic of Esaki tunnel diode [11]

The conditions necessary for tunneling are as follows. (1) There is an occupied energy state on the side where the electron tunnel and the unoccupied energy state exist at the same energy level on the opposite side. (2) The tunneling potential barrier height is low and width is small enough to have a finite tunneling probability. (3) Energy is conserve in the tunneling process. [12]



Figure 1.5 Energy diagram of Esaki diode at each point in I-V characteristic

Fig 1.5 is energy band diagram of the tunnel diode to illustrate the conduction mechanism. (a) At zero bias, fermi level is located on the n-type conduction band and below the valence band of p-type. This case is called degenerate state and there is no current flow. (b) forward bias is applied, the potential barrier is still high and the electron cannot be injected. However, the electrons of the n-region tunnel to the empty state of the p-region valence band. Therefore, a tunnel current occurs. When the forward bias is increased, most of the electrons of the n-region become equal to the energy of the empty state of the valence band of p-region. At this time, a maximum tunneling current is generated. (peak point) (c) As the forward bias continues to increase, the electrons of the n-region, which is the opposite of the empty state of the valence band decrease. Therefore, the tunneling current decreases. (valley point) (d) If more forward bias is applied, electron hole injection occurs through the lower potential barrier, regular diode current occurs. And tunnel current becomes zero. (e) The electrons of the p-region valence band are tunneled into the empty state of the n-region conduction band and generate a large tunneling current. [11, 13, 14]

In this device operating mechanism, it is suitable for high speed rectification due to the electron movement is faster than the others.

1.1.3 Metal-Insulator-Metal tunnel diode

The Metal-Insulator-Metal (MIM) diode is also called a Metal-Oxide-Metal (MOM) diode. The MIM tunneling diode works by quantum electron tunneling in a structure consisting of an ultra-thin insulator film between two metals that induces a low threshold voltage. When the barrier region is very thin, quantum tunneling occurs. If the barrier is very thin, the probability of electron tunneling is increased. This indicates that the MIM tunnel diode is more suitable for high speed operation because tunneling occurs at lower operating voltage than p-n junction diodes and schottky barrier diodes. Therefore, it can be used as a mixer in infrared and optical radiation detectors. [2]



Figure 1.6 SEM image of Cat-whisker Point contact diode [5]

The initially developed MIM tunnel diode was a conventional point-contact-diode. The MIM diode was called a cat-whisker diode. The Cat-whisker diode was tungsten wire that formed a tunnel junction by pressing a metal plate. This type of diode was difficult to fabricate due to some of the dust that formed barrier layers were collected on the surface or some natural oxide layers. And there was a reproducibility problem. Despite these problems, cat-whisker diodes have been fabricated and used in communications for more than 20 years at high frequency rectification of 150 THz at several hertz. MIMI didoes have also been used in image display applications and as switching devices in thin-film transistor liquid crystal display technology (TFT-LCD). [5]

1.1.3.1 Conduction mechanism of MIM diode

The MIM diode has a very thin insulator film structure between two different metal plates. The operation theory of MIM diode is shown in figure 1.7. The work functions of metals are shown as Φ_1 , Φ_2 and the electron affinity is χ . The barrier thickness of insulator is d_t. Here dissimilar metals are used to show asymmetric diodes, but the same metal can be used to form symmetric diodes for each electrode.



Figure 1.7 Energy band diagram of asymmetrical metal MIM diode at zero bias, forward bias, and

reverse bias

(a) At zero bias, the fermi level of metals is aligned. The energy band of the insulator is bent and a built-in field is formed. (b) If negative bias is applied to metal (it can be said that reverse bias is applied to metal 1), direct tunneling can be confirmed. The effective distance of the tunneling barrier can be expressed at d_t , the thickness of insulator. (c) If a larger revers bias is applied, the difference between the fermi levels of the two metals will be larger. And the bent of the insulator will become worse. Therefore, the effective thickness d_t is less than the thickness of the insulator. In this case, the Fowler-Nordheim (FN) tunneling begins. (d) When a metal 1 is positively charged (it can be said that metal 2 has a forward bias), the band shift occurs as shown in (d). In this case, FN tunneling barrier is bent more and the effective thickness d_t becomes shorter, increasing the probability of FN tunneling. [15]



Figure 1.8 Energy band diagram of the direct tunneling and the Fowler-Nordheim tunneling [8]

As shown in figure 1.8, various tunneling processes can be divided in the silicon-insulatorsilicon structure. Depending on the shape of the energy barrier, it can be divided into Fowler-Nordheim (FN) tunneling and direct tunneling. [7] If the voltage across the insulator layer is large enough, the energy band of the tunneling barrier will have a triangular shape, which is FN tunneling. On the other hand, if the voltage across the insulator layer is small, the energy band of the tunneling barrier has a trapezoid, which is direct tunneling. Figure 1.8 is a schematic energy and diagram of direct tunneling. The dielectric energy barrier has a trapezoidal shape. The electrons tunnel through a distance equal to the thickness of the insulator. [7, 16]

Fig1.8 is a schematic energy band diagram of Fowler-Nordheim (FN) tunneling. FN tunneling is a special case of direct tunneling. When enough electric field is applied to the electron to pass through the energy barrier, the electron tunnels the triangular potential barrier. At this time, the electrons tunnel through a distance shorter than the thickness of the insulator, the expression of FN tunneling current is

$$J = \frac{q^{3}E^{2}}{8\pi hq\phi_{B}} \left[\frac{-8\pi (2qm_{T}^{*})^{1/2}}{3hE} \phi_{B}^{3/2} \right],$$

Where q is electronic charge, E is the electric field across the dielectric, h is Planck's constant, $q\phi_B$ is barrier height, m_T^* is the tunneling effective mass in dielectric. [8]

1.1.3.2 Characteristics of MIM diode

MIM diode needs two requirements asymmetrical I-V characteristics and short response time to be an ideal diode.

The asymmetric I-V characteristics are related to the conversion efficiency for rectification performance. The metal electrodes used in MIM tunnel diodes for tunneling must have sufficient work function difference. Figure 1.9 shows the tunnel resistance of an asymmetric structure MIM diode with various thickness of insulator layer; d=20, 30 and 40Å, ϕ_1 =1V and ϕ_2 =2V. The tunneling area of the MIM diode can be explained by the characteristics of the insulating film on the metal substrate. The thinnest part of the insulating film causes the tunneling current flow. This indicates that different materials can make different current flows depending on the voltage. The resistance of the MIM diode should be kept very low. As shown in the figure 1.9, the resistance, across the diode, decreases as the voltage increase. Therefore, the resistance of the diode is very high at low voltage, a thinner insulator is required for effectively affect the quantum tunneling phenomenon. [5]

Another factor is the short response time for high speed rectification. MIM diodes are one of the tunneling device, tunneling transit times must be analyzed to achieve high operating

frequencies.



Figure 1.9 Theoretical tunnel resistance as the applied bias for an asymmetrical MIM structure [5]

When operating at higher frequencies, the device optimized is required to solve low impedance and high non-linearity. The frequency response of the diode is determined by the resistance-capacitance (RC) time constant. The capacitance is proportional to the surface area. The capacitance should be kept as small as possible. Therefore, the device area must be minimized. [2, 5]

1.1.3.3. Theoretical model of MIM diode

Figure 1.10 shows the equivalent circuit of MIM diode, it can be explained the MIM diode model. C_d is a capacitance that it is connected in parallel to the total resistor (R_d), fixed resistor and the non-linear resistor. The internal resistance of MIM diode indicates the fixed resistance and the voltage dependent resistance can be expressed non-linear resistance.



Figure 1.10 Equivalent circuit of MIM diode

In this equivalent circuit, the current flow through the C_d and R_d decreased, where the C_d and R_d should be kept small. Following indicate the cut-off frequency.

$$f_c = \frac{1}{2\pi R_d C_d}$$

Where, f_c is the cut-off frequency of the diode, R_d is total resistance, and C_d is the capacitance of diode. The cut-off frequency is one of the important factors for high speed operation and represents the limited frequency of the diode. As shown in the above equation, to increase the cut-off frequency of the MIM diode, C_d and R_d should be small.

$$C_d = \frac{\varepsilon_0 \varepsilon_r A}{d}$$

This equation expresses the capacitance, where ε_0 is the vacuum permittivity, ε_r is the relative dielectric constant, A is the contact area, and d is the thickness of the insulator. According to the above equation, the capacitance is mainly affected by the thickness of the contact area and the insulator layer, which can be obtained by increasing the increasing the thickness d or decreasing the contact area A.

$$D = e^{-2d\sqrt{\frac{2m(V-E)}{\hbar^2}}}$$

However, according the tunneling probability equation, as shown the above, the probability of tunneling decreases considerably if the non-linearity of the I-V curve of the diode and the thickness of the insulator layer are increased to 5nm or more. Here, D is the transmission probability and d is the thickness of the insulator layer, m is the electron mass, V is the barrier height, and E is the electron energy. According to this equation, the high transmission probability is due to thin insulator layer. Therefore, in order to obtain a lower capacitance, the contact area A should be minimized.

Resistance also affect the high speed operation, it needs to be considered.

$$R_d = \rho \frac{L}{A}$$

Where ρ is the resistivity, L is the length and A is the contact area. The contact area is reduced a small capacitance can be obtained. However, according to the resistance, the small contact area causes a large resistance. In other words, the two parameters are contradictory so that the resistance and capacitance cannot decrease at the same time.

The MIM tunneling diode is a high speed rectification device that is convert alternating current (AC) to direct current (DC). There are factors to be considered when using the MIM diode concept. The parasitic capacitance exists at high frequency. Thus, the MIM device should be designed to minimize parasitic capacitance. Regardless of the external bias, the I-V characteristic of the MIM diode should be non-linear and asymmetry for rectification efficiency. Thus, a zero bias response is needed. To achieve this, MIM diode designed with two metals which have dissimilar work function. [2, 5]

1.1.3.4 Structural tendency of MIM diode

Recently, various types of MIM diodes for high efficiency and high-speed rectification have been studied. For Cat-whisker diodes, which are the initial point contact diodes, use sharp tungsten wire to reduce the contact area. However, the structure is unstable, the structure is changed more stable. Figure 1.11 shows Edge metal-oxide-metal diode. It uses thin native oxide film which is formed a between the flat metal and the vertically bent metal of the edge. This structure is more stable than the point contact structure. With the developing of fabrication technology, a number of devices have been researched which are reduced the size of device and also shows the enhanced characteristics compared MIM diode. Device geometry can contribute in non-linear asymmetric I-V response and show rectification characteristics. [2, 5]



Figure 1.11 The schematic and SEM image of the edge MOM diode [17]



Figure 1.12 The schematic and SEM image of lateral MIM diode [18]



Figure 1.13 The schematic and SEM image of later MIM diode [19]



Figure 1.14 The schematic of bent metal wire MIM diode [20]

1.2 objective

This is the research purpose of the vertical metal cylinder structure MIM diode.

- Non-linearity and asymmetric IV characteristics : To obtain asymmetric IV characteristic, unidirectional current flow, and non-linearity current flow, the selection of materials by work function and electron affinity has to be considered and structure has to be changed.
- Structural design of MIM diode : The difference of the large work function was insufficient to support the effect of the MIM diode. In order to obtain more asymmetric IV characteristics, we had to add a structural effect to the MIM diode. Vertical metal cylinder structure, high aspect ratio, and proper pitch structure have been utilized to increase the field emission effect of MIM diodes. High field emission structure can make more asymmetry IV features.
- Simple process and reproducibility : We have studied a method for easily controlling very thin insulator films tor the tunneling barrier. For direct tunneling and FN tunneling, a dielectric layer with a maximum thickness of 5 nm or less must be maintained. This is one of the most important parts of MIM diodes. In addition, the reproducibility can be improved through a simple process.

II. FABRICATION

We have fabricated three types of MIM diodes, simple flat Metal-Insulator-Metal (MIM) diode, double insulator structure flat Metal-Insulator-Insulator-Metal (MIIM) diode and Vertical Metal Cylinder Structure (VMCS) MIM diode. In this chapter, we will describe in detail the fabrication process of these two devices.

2.1 Fabrication of simple flat MIM diode

Flat MIM diodes were fabricated. This structure was made to compare the structural characteristics of the vertical metal cylinder structure MIM diode. This simple MIM diode was fabricated on Si/SiO₂ (1000 Å) substrate and used to reduce unwanted leakage current. We designed three kinds of samples to investigate the effect of the size of the sample. The contact area of the sampled used in the experiment was $3\mu m \times 3\mu m$, $5\mu m \times 5\mu m$, $8\mu m \times 8\mu m$. Here two different metals, Aluminum (Al), Gold (Au), and Aluminum oxide (Al₂O₃) insulator material were used. The samples prepared were Al-Al₂O₃-Al, and Al-Al₂O₃-Au. Detail information about the sample is provided in Table 2.1.



Figure 2.1 The schematic of the simple flat MIM diode

samples	Material (Bottom electrode-Oxide-Top electrode)	Size of flat area
Sample #1		3μm×3μm
	Al-Al ₂ O ₃ -Al	5μm×5μm
		8μm×8μm
Sample #2		3μm×3μm
	Al-Al ₂ O ₃ -Au	5µm×5µm
		8μm×8μm

Table 2.1 The samples of simple flat MIM diodes



Figure 2.2 The schematic of the fabrication process of the simple flat MIM diode

The Si/SiO₂ substrate was cleaned before preparing the sample. The cleaning process is carried out in acetone and isopropyl alcohol (IPA) for 5 min each with ultrasonic agitation. After the cleaning process, the bottom electrode pattern is formed by photo lithography. The nLOF 2035 negative photo resist (PR) is coated on the Si/SiO₂ substrate by spin coating at 3500 rpm. Then soft baking is done at 110 °C for 60 sec on the hot plate. The wafer was exposed to a UV mercury lamp. The expose wafer is post-soft baked at 110 °C for 60 sec on the hot plate and immersed for 90 sec on the AZ 300 MIF developer. After that, thermal evaporator was used to deposit material on a Si/SiO₂-PR patterned substrate. The material of Al was deposited 100 nm at a rate of 1Å/sec. After deposition, acetone is used to remove PR to form the bottom electrode by lift off process. Lift off can affect how devices work, depend on material, substrate, pattern size, and so on. Since the size of fabricated device is less than 10µm, it can be very sensitive to the process. An oxide layer was formed using an atomic layer deposition (ALD) system. The aluminum oxide (Al₂O₃) dielectric material was formed using plasma enhanced atomic layer deposition (PEALD). Al₂O₃ layer was grown at 200 °C process. Trimethylaluminum (TMA) was used as the precursors for Al₂O₃. The Al₂O₃ was grown by 3 nm thickness. The thickness of the film was confined by an ellipsometer system. Oxide layer was grown and then the pattern was formed and metal was deposited to form a top electrode. The spin coating process is repeated as above and aligned in the photo lithography step so that the structure of bottom electrode and top electrode is well formed. The top metal was also deposited by thermal evaporator and the aluminum (Al) and gold (Au) used as top metal deposition, the lift-off process is performed to form the top electrode.

2.2 Fabrication of flat MIIM diode

Flat Metal-Insulator-Insulator-Metal (MIIM) diode was fabricated to compare rectification effect with MIM diode. This flat MIM diode was fabricated on Si/SiO₂ (1000 Å) substrate and used to reduce unwanted leakage current. We designed also three kinds of samples to investigate the effect of the size of the sample. The contact area of the sampled used in the experiment was $3\mu m \times 3\mu m$, $5\mu m \times 5\mu m$, $8\mu m \times 8\mu m$. Here two different metals; Aluminum (Al), Gold (Au) and two insulator materials; Aluminum oxide (Al₂O₃), Hafnium oxide (HfO₂) were used. The samples prepared were Al-Al₂O₃-HfO₂-Al and Al-Al₂O₃- HfO₂-Au insulator double layer. Figure 2.3 shows the schematic of flat MIIM diode.



Figure 2.3 The schematic of the flat MIIM diode

The fabrication details are almost same with simple flat MIM diode. The figure 2.4 shows the fabrication process details of flat MIIM diode. The samples are fabricated on the Si/SiO₂ substrate. The bottom electrode was pattern by photo lithography and metal was evaporated. The insulator layers, Al₂O₃ and HfO₂, were deposited by PEALD and thermal ALD system to get ultra-thin layers respectively. Each insulator layers have 3 nm thickness. HfO₂ was deposited following Al₂O₃ layer. Trimethylaluminum (TMA) was used as the precursors for Al₂O₃ and Tetrakis (ethylmethlamino) hafnium (TDMAHf) was used as the precursors for HfO₂. The top electrode was pattern by photo lithography aligned with bottom electrode. The top metal was formed by thermal evaporator system and lift off process is performed.

samples	Material (Bottom electrode-Oxide 1-Oxide 2-Top electrode)	Size of flat area
		3μm×3μm
Sample #1	Al-Al ₂ O ₃ -HfO ₂ -Al	5μm×5μm
		8μm×8μm
		3μm×3μm
Sample #2	Al-Al ₂ O ₃ -HfO ₂ -Au	5μm×5μm
		8μm×8μm

Table 2.2 The samples of flat MIIM diodes



Figure 2.4 The schematic of the fabrication process of the flat MIIM diode

2.3 Fabrication of vertical metal cylinder structure MIM diode

We were fabrication the MIM diode using vertical metal cylinder structure on Si/SiO_2 substrate. A substrate of Si/SiO_2 was used to prevent leakage currents flowing through undesired paths. The device was fabricated by mixing photo lithography and electron beam lithography (EBL). The EBL process was used to open the contact pads of the bottom electrode and to get nano-size pattern for small cylinder holes to increase the aspect ratio of the cylinder. EBL is typically used to obtain very small patterns.

Before fabricating the device, use acetone and isopropyl alcohol (IPA) for ultrasonic cleaning for 5 min each. The bottom electrode was formed by photo lithography and deposition of metal. The bottom electrode was formed by the lift off process. The nLOF 2035 negative photo resist (PR) is coated on the Si/SiO₂ substrate by spin coating at 3500 rpm. Then soft baking is done at 110 °C for 60 sec on the hot plate. The wafer was exposed to a UV mercury lamp. The expose wafer is post-soft baked at 110 °C for 60 sec on the hot plate and immersed for 90 sec on the AZ 300 MIF developer. A thermal evaporator was used to deposit metal on a Si/SiO₂-PR patterned substrate. The metal was deposited 100 nm thickness as a rate of 1Å/sec. After deposition, acetone is used to remove PR to form the bottom electrode by lift off process. After forming the bottom electrode, an oxide layer was formed using an atomic layer deposition (ALD) system. The Aluminum oxide (Al₂O₃) dielectric material was formed using plasma enhanced atomic layer deposition (PEALD). Al₂O₃ layer was grown at 200 °C process and Trimethylaluminum (TMA) was used as the precursors for Al₂O₃. The Al₂O₃ layer was grown by 3 nm thickness. The thickness of the film was confirmed by an ellipsometer system. For the electron beam lithography process, a Poly(methyl methacrylate) (PMMA) A4 positive electron beam resistor (ER) is spin coated at a spin rate 5000 rpm and 200 nm thickness PMMA layer was obtained. Then baking at a temperature of 170°C for 5 min. The EBL was carried out using Raith 150^{TWO} Electron beam lithography system. This system was used for patterning with an acceleration voltage of 20kV.



Figure 2.5 The whole devies pattern design for vertical metal cylinder structure MIM diode

EBL was patterned with two designs of hole and pad. The hole and pad exposures of the cylinder were aligned with the bottom electrode. The hole was optimized to 260 pA beam current at 300 mJ/cm² dose with 30 apertures beam. The hole size is 90 nm. And pad was optimized approximately 1100 pA beam current at 250 mJ/cm² dose with 60 apertures beam. The pad size is $90\mu \times 90\mu \times 90\mu m$ rectangular. Develop process was run in 1:3 ratio of methyl isobutyl keptone (MIBK):IPA for 90 sec and then rinsed for 30 sec in IPA. After develop process, shadow mask is aligned for shadow evaporation. The reason why evaporated after by shadow mask alignment, the PMMA polymer layer was used as a supporting between the cylinders and the top electrode and to prevent the direct current path from top electrode to bottom electrode. After the shadow mask alignment, holes were filled with metal to form cylinder structure. The metal was deposited by thermal evaporation the deposition height is 200 nm as a rate of 1Å/sec.



Figure 2.6 The schematic of fabrication process of the VMCS MIM diode

III. ELECTRIC CHARACTERISTICS AND RESULT

3.1 Electrical characteristics of simple flat MIM diode

Simple flat MIM diode has sandwich structure with thin insulator layer between two metal thin films. To confirm tunneling effect and compare with other structure, we fabricated simple flat MIM diodes. It shows tunneling effect, which enables low threshold voltage, high speed switching. The work function difference of two metals makes asymmetric I-V characteristics. The small contact area of metal-insulator-metal leads small capacitance. Then it can be operated as a high frequency diode.



Figure 3.1 Optical microscope image of simple flat MIM diode

Figure 3.1 is an optical microscope (OM) image of simple flat MIM diode. The material setting of MIM diode is Al-Al₂O₃-Al, Al-Al₂O₃-Au and each sample has a contact area of $3\mu m \times 3\mu m$, $5\mu m \times 5\mu m$, $8\mu m \times 8\mu m$. To study asymmetric I-V characteristics, two metals were used with different work function. And to improve the asymmetric I-V, double insulator structure also used.

Metal materials	Work function (ø)
Al	4.18 eV
Au	5.1 eV

Table 3.1 Work funcion of metal materials

Table 3.1 shows the work function of aluminum (Al) and gold (Au). Table 3.2 shows the material characteristic of aluminum oxide (Al₂O₃) and hafnium oxide (HfO₂); electron affinity (χ), energy gap (E_g), dielectric constant (κ).

Table 3.2 The properties of oxide materials [21, 22, 23]

Metal materials	Electron affinity (_X)	Energy gap (Eg)	Dielectric constant (κ)
Al ₂ O ₃	1.3 eV	6.4 eV	7.6
HfO ₂	2.5 eV	5.8 eV	18

In the same metal structure and dissimilar structure, the work function difference is 0 eV and 0.92 eV, respectively. Figure 3.2 is the band diagram of work function difference of MIM with single insulator layer at zero bias.



Figure 3.2 Energy band diagram of the simple MIM diodes at zero bias



Figure 3.3 I-V curves of the Al-Al₂O₃-Al diode

Figure 3.3 shows the I-V characteristics of the MIM structure, Al-Al₂O₃-Al. This currentvoltage (I-V) value is measured by Keithely 4200 SCS in DC probestation with microscope. I-V characteristics are measured at various contact area; $3\mu m \times 3\mu m$, $5\mu m \times 5\mu m$, $8\mu m \times 8\mu m$. The I-V curves are almost symmetry at negative to positive bias. So it is difficult to get high rectifying effect in this samples. Thus, metal material have to be changed which have different work function.

Figure 3.4 is energy band diagrams of Al-Al₂O₃-Al and Al-Al₂O₃-Au at forward bias. The Al-Al₂O₃-Au sample have band bending at zero bias state. Therefore, we expected that asymmetrical I-V characteristics will be improved by using different metal.



Figure 3.4 Energy band diagrams of the Al-Al₂O₃-Al and Al-Al₂O₃-Au diodes at positive bias



Figure 3.5 I-V curves of the Al-Al₂O₃-Au diode by the cross area

Figure 3.5 shows the I-V characteristics of the MIM structure, Al-Al₂O₃-Au. This sample is used the different metal which have different work functions to increase the asymmetric characteristics for high rectifying effect. I-V characteristics are measured at various contact area; $3\mu m \times 3\mu m$, $5\mu m \times 5\mu m$, $8\mu m \times 8\mu m$. Previous page, the asymmetric I-V characteristics are expected in this samples. However, the asymmetric characteristics of I-V curve is still quit small. So it is not easy to obtain high rectifying effect.

Figure 3.6 is I-V characteristics comparing Al-Al₂O₃-Al and Al-Al₂O₃-Au respectively. Samples have 8μ m× 8μ m contact area. As shown in figure 3.2, Al-Al₂O₃-Au samples already have different metal work functions, so the energy band bending is showed at zero bias. When forward bias is applied, the energy band bending is more increase as shown in figure 3.4. So the energy barrier is thinner according to the FN tunneling mechanism and electrons can easily pass through the triangular energy barrier for the tunneling effect. Conversely, when reverse bias is applied, the energy barrier shape becomes more trapezoidal. Therefore, the tunneling pass is increased and the probability of electrons passing through is reduced. So the I-V characteristics are shown more asymmetry. Figure 3.7 is the asymmetric I-V characteristics, η . η is defined as $|I_-/I_+|$, the current at negative bias over the current at positive bias. So if the value is 1, it means perfectly symmetry. To show the plot, there are small increase asymmetry characteristics by using different metal materials which have different work functions. However, it is still hard to show the rectifying effect.

Figure 3.8 shows the Fowler-Nordheim (FN) tunneling model of two samples; Al-Al₂O₃-Al and Al-Al₂O₃-Au. The FN tunneling model is represented by $\ln(I/V_2)$ for 1/V. It can be shown FN tunneling and direct tunneling models at positive bias and negative bias. As shown in this graph, both FN tunneling and direct tunneling can be indicated in all four samples. Especially, the direct tunneling effect is dominant rather than FN tunneling. Direct tunneling is dominant in positive bias. Therefore, the I-V characteristics of the sample are not asymmetry as shown in Figure 3.7. So we have to fabricate other structure of MIM diode.



Figure 3.6 I-V curves of the comparing with Al-Al₂O₃-Al and Al-Al₂O₃-Au diodes



Figure 3.7 Asymmetry plots of the comparing with Al-Al₂O₃-Al and Al-Al₂O₃-Au diodes



Figure 3.8 FN tunneling model of Al-Al₂O₃-Al and Al-Al₂O₃-Au diodes at postivie bias

3.2 Electrical characteristics of flat MIIM diode

Previous experiments, the asymmetric I-V characteristics are not obtained. So we fabricate other structure of MIM diode.



Figure 3.9 Energy band diagram of simple flat MIIM diode at zero bias

To increase the asymmetrical characteristics and to obtain high rectification effect, double insulator structure, MIIM diode was studied. [24]

Figure 3.9 is the band diagram of work function difference of MIIM with double insulator layer at zero bias. Figure 3.10 and figure 3.11 show the band diagram of MIIM diode, Al-Al₂O₃-HfO₂-Al and Al-Al₂O₃-HfO₂-Au at forward and reverse bias.



Figure 3.10 Energy band diagram of simple flat MIIM diode, Al-Al₂O₃-HfO₂-Al



Figure 3.11 Energy band diagram of simple flat MIIM diode, Al-Al₂O₃-HfO₂-Au

Figure 3.12 shows the I-V curves of two MIIM diodes, Al-Al₂O₃-HfO₂-Al and Al-Al₂O₃-HfO₂-Au. I-V characteristics of Al-Al₂O₃-HfO₂-Al MIIM diode are asymmetry. The band diagram of this sample is indicated in figure 3.10. At positive bias, the energy band is bent. Then the thickness of the tunneling barrier becomes thinner than the thickness at positive bias. As a result, electron tunneling probability is increase and tunneling is occur easily. Therefore, a large current flows when a positive bias. On the other hand, in negative bias, the energy band bending also occurs at negative bias. However, the tunneling barrier thickness is same with the insulator thickness. Therefore, the electrons should pass through two insulator tunneling barriers, as shown in the figure 3.10. Thus, the devices show asymmetric I-V characteristic with higher current flow at positive bias than negative bias.

Figure 3.12 also shows the I-V curve of Al-Al₂O₃-HfO₂-Au. Unlike the Al-Al₂O₃-HfO₂-Al, this MIIM diode has no asymmetrical I-V curves. In the above band diagram, figure 3.11, the band bending exhibits similar at positive and negative bias. So, the tunneling barrier thickness is same with the insulator thickness at two bias state. In this case, electron tunneling occurs in both insulator layers. Therefore, this MIIM diode shows a symmetric I-V characteristic.



Figure 3.12 I-V curves of the Al-Al₂O₃-HfO₂-Al and Al-Al₂O₃-HfO₂-Au diodes



Figure 3.13 I-V curves of the comparing with Al-Al₂O₃-HfO₂-Al and Al-Al₂O₃-HfO₂-Au diodes



Figure 3.14 Asymmetry plots of the comparing with Al-Al₂O₃-HfO₂-Al and Al-Al₂O₃-HfO₂-Au diodes



Figure 3.15 FN tunneling model of Al-Al₂O₃-HfO₂-Al and Al-Al₂O₃-HfO₂-Au diodes at positive bias

The comparison of I-V characteristics of the two samples, all contact area is $8\mu m \times 8\mu m$, are shown in figure 3.13. Al-Al₂O₃-HfO₂-Al diode has high asymmetry and high current level than Al-Al₂O₃-HfO₂-Au diode. Figure 3.14 is shown the asymmetry characteristics of the two samples. I-V asymmetry, η , is defined as $|I_-/I_+|$ and the $\eta=1$ represents symmetry. The η value of Al-Al₂O₃-HfO₂-Al is approximately 0.175 and the η value of Al-Al₂O₃-HfO₂-Au is approximately 0.957 at 2V. So the Al-Al₂O₃-HfO₂-Al diode represents high asymmetric I-V characteristics.

Figure 3.15 shows the FN tunneling model of two MIIM diodes at positive bias. In this model, the result of Al-Al₂O₃-HfO₂-Al MIIM diode is shown both FN tunneling and direct tunneling characteristics. However, the FN tunneling is dominant then direct tunneling. On the other hand, Al-Al₂O₃-HfO₂-Au MIIM diode has almost no FN tunneling properties but direct tunneling is dominant. This result is consistent with the previous energy band diagrams and I-V curves. Therefore, it needs proper material selection with double insulator layer structure.



Figure 3.16 Schematic of flat MIM diode design for RF measurement

In the future, this flat MIM and MIIM tunnel diode can be studied at radio frequency (RF) for high-speed switching. Due to the low series resistance, the MIM didoes are expected to operate at high frequency. [26] Figure 3.16 is the schematic of the MIM diode design for RF measurement system. This MIM diode will be fabricated by photo lithography and ALD, metal evaporation. Same materials will be used Al, Au, and Al₂O₃. The coupling of the MIM diode is through RF probe and the Alternating Current (AC) signal will be generated by network analyzer. The S-parameter can be measured. After we measure the S-parameter using network analyzer, we can get output DC voltage as a function of input RF power and cut-off frequency. And we can get the effective efficiency that is defined as the ratio of output DC power to the available input RF power.

3.3 Electrical characteristics of vertical metal cylinder structure MIM diode

Metal-Insulator-Metal (MIM) diode using vertical metal cylinder structure (VMCS) has a metal cylinder array between two metals. The holes for cylinder were formed by the electron beam lithography (EBL) to form small size hole for high aspect ratio cylinder and the cylinder metal was easily filled by evaporation system. To reduce the current, flowing directly between the top electrode and the bottom electrode, PMMA was used as the supporting layer. So the current flow Metal-insulator-metal cylinder-metal pathway. And the tip of the vertical metal cylinder has a high electric field density. The vertical metal cylinder structure has a simple process by forming through EBL and evaporator system. It is a diode that has high reproducibility than Carbon Nanotubes (CNTs), metal nano-rods and nanowires, which are conventional vertical metallic material. This VMCS MIM diode can easily controlled the deposition of ultra-thin insulator layer compared with the lateral MIM diode. It was fabricated to improve the non-linearity and asymmetric I-V characteristics of the MIM diode by changing the structure of the MIM diode. And it shows tunneling effect, which enables low threshold voltage, high speed switching.



Figure 3.17 The schematic of VMCS MIM diode



Figure 3.18 Optical microscope and SEM image of the VMCS diode and cylinder array

We fabricate VMCS diode, Al-Al₂O₃-Al. Aluminum (Al) is used as cylinder and top electrode metal and Aluminum oxide (Al₂O₃) is used as insulator layer for tunneling layer. Figure 3.18 is an optical microscope (OM) image and SEM image of the devices. The cylinder arrays are well formed and the top electrode-cylinder is contacted well. The diameter of the cylinder is 100 nm and the height is 200 nm. So, the cylinder has 2 aspect ratio.



Figure 3.19 J-V curve of VMCS MIM diode with Al metal (Inset is I-V plot)

Figure 3.19 shows the J-V characteristics of VMCS diodes with Al metal and the inset graph is I-V characteristics of the samples. The metal cylinder arrays are 30×30 and 35×35 arrays, and the total areas of the cylinder arrays contact with metal-insulator are 7.065×10^{-10} cm² and 9.616×10^{-10} cm² each. However, the current densities are too low both cylinder array samples. Because, the cylinders are not fully filled-up by evaporation. So the vacuum layer occurs in the cylinder.

Figure 3.20 is the top view of the cylinder. In this SEM image, the cylinder was not completely filled, and it looks like the donut shape hollowed at the center. Therefore, the device has low current flow. So in order to solve this problem, the metal was changed to gold (Au).



Figure 3.20 SEM images of Al metal cylinder at the top view

In order to solve the problem that the complete cylinder was not formed, the metal was replaced with Au. The insulator material is same and the device fabrication process is also same.

Figure 3.21 shows the SEM images of the Au metal cylinder viewed from the side (45 ° tiled) and top. The hole size of cylinder is 100 nm and has a pitch of 1 μ m. The height of the cylinder is 200 nm and has 2 aspect ratio same with the previous sample, Al metal cylinder. It can be confirmed that the cylinder is completely filled circle.

Figure 3.22 shows the J-V characteristics of the VMCS MIM diode with Au metal, inset is I-V curves. J-V characteristics show almost symmetrically. It shows high current flow compare with Al metal cylinder structure.



Figure 3.21 SEM images of Au metal cylinder height and diameter



Figure 3.22 J-V curve of VMCS MIM diode with Au metal cylinder



Figure 3.23 Comparison the asymmetry of the VMCS diode with flat MIM diode



Figure 3.24 FN tunneling model of VMCS MIM diode with Au metal cylinder at positive bias (Inset is FN characteristic at negative bias)

The asymmetry of VMCS MIM diode is shown in figure 3.23. It is compared with the asymmetry characteristics of simple flat MIM diodes. To show the graph, the η value of the VMCS MIM diode is 0.68 and the both values of the simple flat MIM diodes is approximately 1. So the asymmetry characteristic is increased than the simple flat MIM diodes.

The FN tunneling model is shown in figure 3.24. It shows both FN tunneling and direct tunneling, at positive bias and negative bias. This indicate that the MIM diode using vertical metal cylinder structure operate with a tunneling mechanism at low voltage, 1V. Therefore, the cylinder structure can affect the tunneling mechanism.

After we show the structural effect of VMCS, we studied to increase the electrical properties for rectifying effect. So we change the cylinder structure array pitch and aspect ratio. We expect to increase cylinder pitch then the current level also increase. Because, the local electric field is focused on the cylinder structure.





Figure 3.25 is SEM images of the vertical metal cylinder arrays which have different pitch. The pitch is 200 nm, 400 nm, and 1 μ m each and the diameter of the hole is 100 nm. And the table 3.3 is shown the real hole diameter of the cylinder structure.

Pitch	200 nm	400 nm	1 μm
Real diameter	120 nm	108 nm	100 nm

Table 3.3 Real hole diameter	r of the cyilnder structure
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Figure 3.26 J-V characteristics of VMCS MIM diodes with different cylinder array pitch

The J-V characteristic of the VMCS MIM diode with different cylinder array pitch is shown in figure 3.26. We expect to increase the current level when increase the cylinder array pitch. However, to show the J-V characteristics, the current density is decrease when the cylinder pitch is increase. Because, due to the electron beam lithography overlay, the real hole size is different with designed pattern. So decrease the cylinder array pitch, the real hole size is increase. Therefore, the current density is also increase at small cylinder array pitch.



Figure 3.27 SEM images of 50 nm hole diameter cylinder array



Figure 3.28 J-V characteristic of 4 aspect ratio cylinder MIM diode



Figure 3.29 Asymmetry of the VMCS diode with 4 aspect ratio cylinder

And we fabricated the 4 aspect ratio vertical metal cylinder structure for MIM diode to increase the cylinder structure effect for rectifying effect. The cylinder hole size is 50 nm and the height is 200 nm. So the aspect ratio of the cylinder is 4.

Figure 3.27 is shown the vertical metal cylinder arrays which have 50 nm hole diameter. To show the J-V characteristic in figure 3.28, the current density is slightly decreased than 2 aspect ratio VMCS MIM diode. However, to compare the two samples, the asymmetry characteristic is increased when the cylinder aspect ratio is increase than 2 aspect ratio case as shown in figure 3.29. Therefore, the aspect ratio of the cylinder is increase the asymmetry characteristic for rectifying effect is also increase. As a result, we can show that the structure of the MIM diode can be affected the asymmetric electrical properties and tunneling mechanism.

IV. CONCLUSION

Simple flat MIM, MIIM diode, vertical metal structure MIM diode have been fabricated and studier the electrical characteristics for high rectification.

The properties of simple flat MIM diode operated by tunneling mechanism was studied and the structural effect of MIM tunneling diode was studied by fabricating vertical metal cylinder array structure. The diode was fabricated by simple process compared with other device using vertical CNTs, nano-rods, and nano-wires and the reproducibility was improved. In addition, the diode has been studied to improve the electrical characteristics, non-linearity and asymmetry, for high rectifying effect. The energy band diagram and the FN tunneling model were used to characterize the device.

The simple flat MIM diodes show high current using ultra-thin insulator layer by ALD system. However, the MIM diode that have same metal and work function show almost linear and symmetry I-V characteristics. Therefore, it was not suitable for rectification. So the other flat MIM diode was fabricated that have different metal and work functions difference. However, this diode also lacked non-linearity and asymmetric properties. And the simple flat MIM diodes indicate direct tunneling mechanism dominantly. The MIIM diode using double insulator layers show the I-V characteristics to get more high non-linearity and asymmetry. The asymmetry, η , was calculated to be 0.166. Therefore, the MIIM diode can have high rectification for rectifying diode.

In order to improve the rectifying effect, the VMCS MIM diode was studied using EBL system. This VMCS diode can control the tunneling layer gap more easily than lateral MIM diode. And it is expected to have similar structural effect on MIM tunneling diode. However, VMCS MIM diode also showed little non-linearity and little asymmetric I-V characteristics. The device shows similar current flow tendency with simple flat MIM diode. Therefore, this diode showed similar with simple flat MIM diode that has very small contact area. But, FN tunneling model shows more FN tunneling mechanism than direct tunneling in VMCS MIM diode compared with simple flat MIM diode. It can be seen that the cylinder structure affects the slope of the tunneling energy barrier and can affect the I-V characteristics of the device. In other words, the asymmetry and non-linear characteristics of the VMCS MIM diode are weak, but the diode is operated by the tunneling mechanism and the diode has structural effect. Therefore, that can be lead to improve the characteristic of VMCS MIM diode that are designing high aspect ratio of metal cylinder to increase the structural effect and using dissimilar metal that have different work function.

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요약문

정류 효과를 위한 다양한 재료 및 구조적 디자인의 금속-절연체-금속 터널 다이오드

초고속 동작 및 정류 효과를 위하여 다양한 물질과 구조적인 디자인을 이용한 금속-절연체-금속 (MIM) 터널 다이오드의 전기적인 특성이 연구되어지고 있다. 최근 초고속 소자는 고성능 CPU, 광전기적 소자 그리고 통신과 같은 많은 영역에서 요구되어진다. 고속 동작을 위한 다양한 전기적 성분 중, 본 논문에서는 고속 소자를 위한 기본적인 구성요소인 정류 다이오드에 대하여 연구하였다.

전통적으로, pn 접합 다이오드와 쇼트키 다이오드가 정류 효과를 위해 집중적으로 연구되어졌다. 그러나, pn 접합 다이오드의 작동 메커니즘은 큰 공핍 영역 때문에 고속 정류소자로는 적합하지 않다. 쇼트키 장벽 다이오드의 경우, 매우 짧은 역 회복 시간을 가지기 때문에 고속 동작에 적합하다. 하지만, 쇼트키 다이오드의 작동 주파수가 5THz 로 제한이 된다. 그러므로 높은 주파수 범위에서 작동하는 새로운 정류 소자에 대한 연구가 필요하다. 정류 다이오드 중 하나인 MIM 다이오드는, 입자가 매우 얇은 포텐셜 장벽을 통과할 때 발생하는 터널링 메커니즘에 따라 작동한다. 따라서 MIM 터널 다이오드는 고속 스위칭이 가능하고 낮은 작동 전압을 가진다.

MIM 다이오드 구조의 고속 작동과 정류 효과를 증가시키기 위해서, MIM 다이오드는 터널링 장벽으로 매우 얇은 절연막을 가져야한다. 대부분 소자의 전류-전압 특성은 비 선형성과 비 대칭성을 가져야만 한다. 소자의 터널링 메커니즘에 의해서 비선형성과 비대칭적 특성이 향상 되어야한다. 따라서 본 논문에서는 MIM 다이오드의 전류-전압 특성의 비선형성과 비대칭성을 향상 시키는 것에 초점을 두었다. 다이오드의 비선형성과 비 대칭성 특성을 향상시키기 위해, MIM 다이오드는 다른 일 함수를 갖는 금속, 두 층의 절연막 그리고 소자의 구조가 중요한 변수로 작용하였다. Simple flat MIM 다이오드, 금속-절연체-절연체-금속의 이중 절연막 구조를 갖는 MIIM 다이오드 그리고 수직적인 금속 실린더 구조를 갖는 MIM 다이오드에 대해 연구하였다. 물질적인 효과를 고려한 simple flat MIIM 다이오드는 비선형성과 비대칭성 특성을 거의 나타내지 않는다. 그러나 flat MIIM 다이오드는 전류-전압 특성에서 좋은 비선형성, 비대칭성 특성을 나타내었다. 수직 금속 실린더 구조의 MIM 다이오드는 비선형성과 비대칭성은

53

부족하지만, simple flat MIM 다이오드에 비해 향상된 비대칭성과 비선형성 특성을 확인 할 수 있었다. 따라서 소자의 구조가 중요한 변수로 적용된 수직 금속 실린더 구조를 갖는 MIM 다이오드는 정류 및 고속 동작으로의 실현가능성을 보여준다. 이러한 새로운 MIM 다이오드는 다양한 새로운 전자소자, 에너지 하베스팅을 위한 정류 안테나, 광학 소자, 고 주파수 영역에서의 적외선/테라 헤르츠 검출기 그리고 전기적인 고속 스위치 소자 등 다양한 분야에서 응용 될 수 있다.

핵심어 : MIM 다이오드, MIIM 다이오드, 수직 금속 실린더 구조, 비선형성, 비대칭성