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P10-1

MFIS-structure Memory Device with High Quality Ferroelectric $Sr_2(Ta_{1-x}Nb_x)_2O_7$ Formed by Physical Vapor Deposition and Oxygen Radical Treatment by Radical Oxygen Assisted Layer by Layer(ROALL) deposition

Hiroyuki Sakurai, Ichirou Takahashi, Tatsunori Isogai, Kiyoshi Funaiwa, Tomoya Tsunoda, Tetsuya Goto, Masaki Hirayama, Akinobu Teramoto,Shigetoshi Sugawa, and Tadahiro Ohmi

New Industry Creation Hatchery Center, Tohoku University
Aza Aoba 04 , Aramaki , Aoba-ku, Sendai, 980-8579, Japan
Phone:+81-22-217-3977 Fax:+81-22-217-3586 e-mail:h-sakurai@fff.niche.tohoku.ac.jp

1. Introduction

A ferroelectric memory is one of the most promising non-volatile memory for low power consumption , high endurance and high speed writing. Recently, ferroelectric field effect transistor such as metal-ferroelectric-insulator-silicon (MFIS-FET) and metal-ferroelectric-metal-insulator-silicon (MFMIS-FET) [1] [2] have attracted much attention from the viewpoints of nondestructive readout and high density memery LSI in accord with the ULSI scaling rule.

In this paper, we demonstrate high-quality ferroelectric $Sr_2(Ta_{1-x}Nb_x)_2O_7$ (STN) film formation process with excellent data retention property more than 1 week even STN film thickness was 100nm fabricated by way of Plasma PVD method. The ferroelectirc films were oxidized by oxygen radical generated from krypton and oxygen gases using microwave-exited(2.45GHz), high density more than 10^{12} cm^{-3}, low electron temperature less than 1eV plasma equipment with the radial line slot antenna.[3]

2. Experiment

Radical Oxygen Assisted Layer by Layer(ROALL) deposition Method

The 10nm of STN film was fabricated on SiO_2/Si substrate by RF-magnetron sputtering(13.56MHz) under Kr/O_2 ambient. The film was strongly oxidized at 400℃ for 60min by oxygen radical using microwave-exited high density more than $10^{12}$$cm^{-3}$, low electron temperature less than 1eV plasma equipment with the radial line slot antenna. . The oxygen radical treated STN was crystallized at 950℃ 60min as a initial crystal. The 10nm of STN was deposited on the initially crystallized STN(10nm). The 20nm(10nm+10nm) of STN film were strongly oxidized again by oxygen radical for 30min using microwave-exited high density plasma. The process of STN deposition and oxygen radical oxidation were repeated until final STN thickness reach to 100nm .Oxygen radical treated STN films were crystallized at 950℃ for 90min. Iridium oxide was deposited on the STN film by RF-magnetron sputter under Xe/O_2 ambient. as upper electrode. To investigate effect of oxygen radical oxidation, the number of oxygen radical treatment were changed. The process flow of MFIS structure was shown in Fig.1.

3. Results and Discussions

The ionized energy of Sr, Ta, Nb, Si shown in Table.1 indicates that it is difficult to ionize Ta^0 and Nb^0 to pentavalent Ta^{5+} 126.2eV/atom and Nb^{5+} 134.0eV/atom compared to Sr^{2+} (16.7eV/atom) and Si^{4+} (103.1eV/atom).

Fully oxidized thickness of Sr Ta Nb were estimated by ESCA analysis as shown in Fig.2 . ESCA chart exhibits the oxidized depth of Nb and Ta were about 10nm and more than 20nm respectively. The 10nm of STN each layer was selected to oxidize sufficiently. On the contrary, oxygen couldn't introduce to Nb with molecular oxygen, nevertheless annealed at 950℃ for 10hrs as previous work reported[4].

The XRD patterns of STN/SiO_2 film annealed at 950℃ was shown in Fig. 3. This result indicated the 100nm of initial STN layer was crystallized. Fig.4 indicated the reciprocal mapping of STN film exhibits(010) and (011) axis orientation.

The C-V characteristics and leakage current density of oxygen radical treatment dependence were indicated Fig.5 and Fig.6 respectively. In case ten times oxygen radical treatment, 0.8V memory window was observed when applied voltage was swept between –5V and 5V. In addition to the ferroelectric memory window, oxygen radical treatment diminish leakage current density of MFIS structure. Ten times oxygen radical treatment improved more than one order of leakage current density than that of two times.

The result of retention property of MFIS structure was shown in Fig.7 . The retention property written voltage are –5V and 5V, and held at 0V described capacitances were kept more than 1 week. These results insisted oxygen radical introduced to STN layer effectively.

4.Conclusions

We demonstrate step by step process STN exhibit 0.8V of ferroelectric memory window when swept between –5V and 5V. The retention property written voltage are –5V and 5V, and held at 0V describes capacitances were kept more than 1 week. These results insist effectively oxidized by oxygen radical ferroelectric film demonstrate excellent ferroelectric properties.

5. References

[1] Hirai T, Y.Tarui, Jpn. Appl.Phys.,**33**(1994) 5219.
[2] H.Takasu, U.S Patent 5,307,305 (1992)
[3] K.Sekine, T.Ohmi et al., IEEE T-ED., **48**(2001) 1556.
[4] I.Takahashi, T. Ohmi et al., ISIF., 11-09-C (2004)

1) O* Treatment Ten times Process

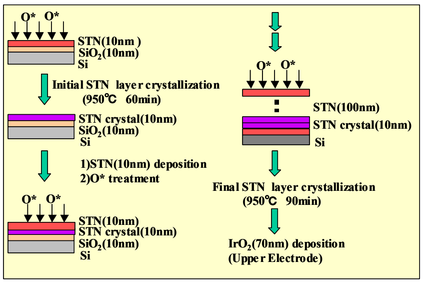
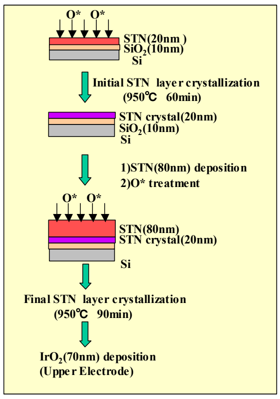


Fig.1 The process flow of MFIS Structure by Radical Oxygen Assisted Layer by Layer (ROALL) method
1)O* Radical Treatment 10times 2) 2times

2) O* Treatment Two times Process



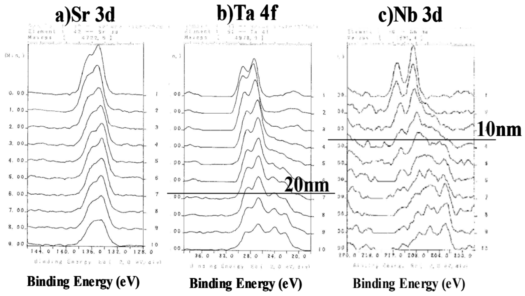


Fig.2 Depth Profiles of STN film by ESCA analysis

O*radical treatment conditions: Power:2.0kW, Substrate temp. :400℃, Kr/O2: 1Torr. ESCA analysis was carried out by ESCA 1000
b)Ta was oxidized nearly 20nm depth. c) Nb was oxidized nearly 10nm depth by Oxygen radical treatment.

Table.1 Ionized energy of metal element

(eV/atom)

	Order					total
	1	2	3	4	5	
Sr	5.7	11.0	-	-	-	16.7
Nb	7.9	16.2	22.3	33.1	46.8	126.2
Ta	6.9	14.3	25.0	38.3	49.4	134.0
Si	8.1	16.3	33.5	45.1	-	103.1

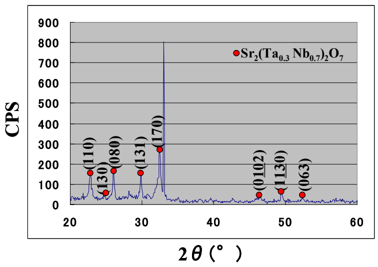


Fig.3 XRD pattern of $Sr_2(Ta_{0.3}Nb_{0.7})_2O_7$(STN) film fabricated by ROALL method (as shown in Fig.1). Oxygen radical were introduced to each 10nm STN layer(10 times treatment).

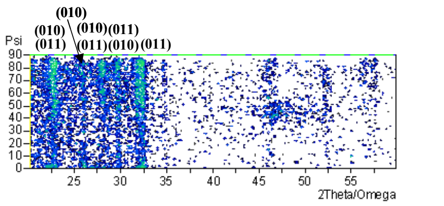


Fig.4 The reciprocal mapping of ROALL process STN/SiO_2/Si substrate. Oxygen radical were introduced to each 10nm STN layer (10 times treatment).

O* treatment 10times

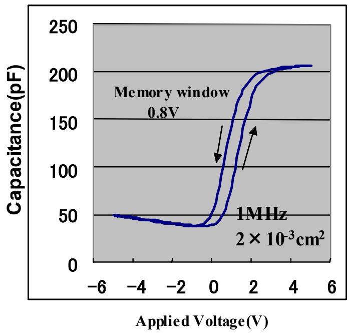
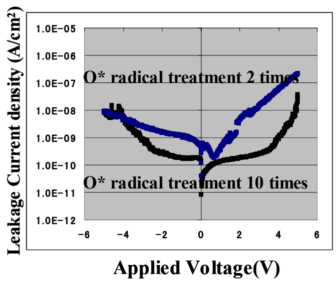


Fig.5 C-V characteristics MFIS structure.
O*radical treatment 10times.
Process flow of MFIS structures was shown in Fig.1.

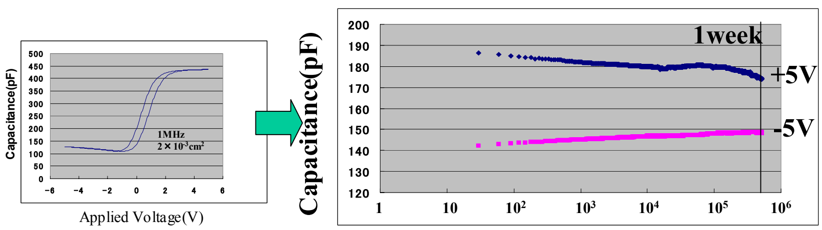


Leakage Current Density

	O* radical treatment	
	2 times	10 times
0.05V	5.2E-10	1.7E-11
1.0V	4.0E-10	9.2E-11
3.0V	1.5E -8	2.5E-11

(A/cm²)

Fig. 6 Leakage Current Density of MFIS structure fabricated by O*radical treatment process



Retention Time (sec) Writing Voltage:±5V
Hold Voltage : 0V

Fig.7 Capacitance retention characteristics of ROALL method MFIS structure