Charging Effect of Aluminum Nitride Thin Films Containing Al Nanocrystals

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In this work, the Al-rich AlN thin film is deposited on Si substrate by radio frequency (RF) sputtering to form a metal-insulator-semiconductor (MIS) structure. Al nanocrystals (nc-Al) are formed and embedded in the AlN thin film. Charge trapping/detrapping in the nc-Al leads to a shift in the flat-band voltage ($V_{FB}$) of the MIS structure. The charge storage ability of the AlN thin films containing Al nanocrystals provides the possibility of memory applications. On the other hand, charge trapping in nc-Al reduces the current conduction because of the breaking of some tunneling paths due to Coulomb blockade effect and the current conduction evolves with a trend towards one-dimensional transport.

Keywords: Al Nanocrystal, Charging Effect.

1. INTRODUCTION

Aluminum nitride (AlN) thin film has attracted much research attention because it is a promising material for applications in surface acoustic wave (SAW) devices and light-emission devices.1–3 In addition, AlN also has an application potential in the area of solar cell synthesis. Many techniques can be used to synthesize AlN films such as reactive evaporation, magnetron sputtering, pulsed laser deposition, and hybrid techniques such as plasma- or ion-assisted depositions.4–7 With its high thermal conductivity,8 reasonable thermal match to semiconductors (such as Si, GaAs and GaN) and small lattice mismatch8,9 and wide band gap (6.2 eV),10 AlN thin film could be used as a gate dielectric for field-effect transistors. In this work, the Al-rich AlN thin film is deposited on Si substrate by radio frequency (RF) sputtering to form a metal-insulator-semiconductor (MIS) structure. Al nanocrystals (nc-Al) are formed and embedded in the AlN thin film. Charge trapping/detrapping in the nc-Al leads to a shift in the flat-band voltage ($V_{FB}$) of the MIS structure. The situation is similar to the memory effect observed for SiO$_2$ thin films containing Si nanocrystals (nc-Si), which has already been used for nonvolatile memory applications.11,12

The charge storage ability of the AlN thin films containing Al nanocrystals provides the possibility of memory applications with low cost. On the other hand, the charging effect on the current conduction in AlN films has been studied also. It is found that charging in the nc-Al leads to a reduction in the current conduction, and the system evolves with a trend towards one-dimensional transport, which has been defined in Ref. [13]. The phenomena can be explained by a model of charge transport in nc-Al arrays.

2. EXPERIMENTAL DETAILS

A series of Al-rich AlN films were deposited on $n$ or $p$-type, (100) oriented Si wafers. Depositions were carried out by RF magnetron sputtering of a pure Al target in a gas mixture of argon and nitrogen. The purities of the argon and nitrogen gas are more than 99.99 in percentage. The flow rates of Ar and N$_2$ were varied to obtain different Al to N ratios. A 200 nm aluminum layer was then deposited to form the gate electrode. The wafer backside was coated with a layer of aluminum with a thickness of about 500 nm after removing the initial oxide. Finally, an alloying process was conducted at 425 °C in N$_2$ ambient to form ohmic contacts. X-ray photoemission spectroscopy (XPS) analysis was performed using a Kratos AXIS spectrometer with monochromatic Al Kα (1486.71 eV) X-ray radiation. After the correction with the appropriate relative sensitivity factors, the Al and N atomic ratios were determined from the calculation of the ratio of $I_{Al}/I_{N}$ where $I_{Al}$ is the peak area of the Al 2p peak.
and $I_{NI}$ is the peak area of the N 1s peak. As the ratio is larger than 1, ranging from 1.2 to 3.5, the as-fabricated films are rich of aluminum. Depth XPS analysis was also carried out. There is no obvious difference in the Al:N ratio at various depths. Transmission Electron Microscopy (TEM) image shows that excess Al atoms form nano-size crystals in the AlN matrix as illustrated in the inset in Figure 1(a), which is also presented in Refs. [14, 15].

Figure 1(a), which is also presented in Refs. [14, 15]. In air environment, the experiment of charging/discharging the Al nanocrystals and the current–voltage ($I–V$) measurements were carried out with a Keithley 4200 semiconductor characterization system, and capacitance–voltage ($C–V$) measurements were carried out with a HP4284A LCR meter at the frequency of 1 MHz.

3. RESULTS AND DISCUSSION

Figure 1(a) shows the $C–V$ characteristics obtained by sweeping the voltage from $−8 \text{ V}$ to $2 \text{ V}$ first and then from $2 \text{ V}$ back to $−8 \text{ V}$. A flat-band voltage shift of $∼1.1 \text{ V}$ is observed from Figure 1(a). The large flat-band voltage shift indicates significant charge trapping in the Al-rich AlN film. The charge trapping is concerned with the Al nanocrystals embedded in the AlN thin films, and the situation here is very similar to that of Si-rich SiO$_2$ (i.e., SiO$_{1−x}$, $x < 2$) thin films where charge trapping in the Si nanocrystals leads to a flat-band voltage shift$^{11,12}$.

Both positive and negative charge trapping in the Al nanocrystals are possible, depending on the gate voltage applied. As shown in Figure 1(b), the application of $30 \text{ V}$ to the gate for 0.1 s shifts the $C–V$ characteristic to the positive with a flat-band voltage shift of $∼1.8 \text{ V}$ (i.e., $\Delta V_{FB} ≈ +1.8 \text{ V}$) indicating a negative charge trapping in the Al nanocrystals. In contrast, as shown in Figure 1(c), the application of $−23 \text{ V}$ for 0.1 s leads to a negative flat-band voltage shift of about 1.3 V (i.e., $\Delta V_{FB} ≈ −1.3 \text{ V}$), indicating positive charge trapping in the Al nanocrystals. These results show that the charge trapping depends on the voltage applied. The capability of charge storage in the Al nanocrystals and the large effect on the flat-band voltage shift provide the possibility of the application of the Al-rich AlN thin films in memory devices.

Figure 2(a) shows flat-band voltage shift as a function of writing time at various voltages. The flat-band voltage shift is increased with the writing time. The application of a higher voltage also leads to a higher flat-band voltage shift. This is because under a positive gate voltage, electrons are injected from the substrate and then trapped in nc-Al. A longer writing time or a higher voltage causes more electron trapping in nc-Al, and thus a larger flat-band voltage shift is observed. For a charging duration that is long enough, for example 100 seconds at 20 V, the flat-band voltage shift tends to saturate, because most of nc-Al are charged up in this situation. This is not shown in the figure, because a writing duration of 100 s is too long for memory applications. On the other hand, there is no shift for $t ≤ 10^4 \text{ s}$, which means that only a voltage application longer than $10^4 \text{ s}$ can write the device. Figure 2(b) shows the retention study of the MIS device. Initially, an application of $+25 \text{ V}$ for 0.1 s was used to write the device. As can be seen in the figure, the flat-band voltage shift is reduced from $∼1.6 \text{ V}$ to $∼1.0 \text{ V}$ after 24 hours, and it is predicted that the flat-band voltage shift will be further reduced to $∼0.5 \text{ V}$ after one year.
respectively. Equation (1) can be used to fit our experimental result of the system of nc-Al embedded in AlN matrix, and the fitting can yield the values of the factors including $\zeta$, $V_{th}$, and $I_0$ of Eq. (1). Figure 3 shows such a fitting to the $I-V$ characteristics before and after application of $+15$ V for 100 s. The $I-V$ characteristics before and after application of $+15$ V for 100 s are shown in (a) and (b), respectively. The lines show the fittings based on Eq. (1). The fittings yield $I_0 = 3.11 \times 10^{-10}$ A and $\zeta = 1.93$ for the situation before the voltage application and $I_0 = 6.00 \times 10^{-11}$ and $\zeta = 1.13$ for the situation after the voltage application. The $\zeta$ value ($=1.93$) for the virgin situation exceeds the two-dimensional transport limitation (i.e., $\zeta = 5/3$). This indicates that the current conduction of the system is beyond the regime of two-dimensional transport. We believe some three-dimensional transport exists in the system in addition to the one- and two-dimensional transport for the virgin situation. After charging by $+15$ V for 100 s, the $\zeta$ is reduced to 1.13 falling in the range of 1–5/3, indicating that the current conduction of the system becomes a quasi two-dimensional transport (i.e., in between one- and two-dimensional transport). The situation could be like that there are two-dimensional tunneling paths in addition to one-dimensional tunneling paths. For both cases, the threshold voltage is found to be approximately zero at room temperature.

As can be observed from Figure 4, the charge trapping leads to a reduction in $\zeta$. $\zeta$ decreases from 1.93 for charging time $t = 0$ (i.e., the virgin case) to 1.13 for charging time $t = 0$ (i.e., the virgin case) to 1.13

$$I = I_0(V - V_{th})^\zeta$$

where $\zeta$ is the scaling exponent, and $V_{th}$ is the threshold voltage. The scaling exponent $\zeta = 1$ and 5/3 for the one- and two-dimensional arrays of quantum dots, respectively. Equation (1) can be used to fit our experimental result of the system of nc-Al embedded in AlN matrix, and the fitting can yield the values of the factors including $\zeta$, $V_{th}$, and $I_0$ of Eq. (1). Figure 3 shows such a fitting to the $I-V$ characteristics before and after application of $+15$ V for 100 s. $\zeta$ is found to be larger than 5/3 for the virgin situation, but it reduces to the range of 1–5/3 after charging by $+15$ V for 100 s.
for $t = 100$ s. It means that it starts from a value bigger than the upper limit ($\zeta = 5/3$) and drops in the range between the upper limit and the lower limit ($\zeta = 1$). This indicates that the current conduction of the system evolves from a situation beyond two-dimensional transport (some three-dimensional transport in the system) to a quasi two-dimensional transport. The evolution has a trend towards the one-dimension transport when the charge trapping is increased.\textsuperscript{13,15} This evolution is due to the breaking of some tunneling paths as a result of charge trapping in the nc-Al, as discussed below.

Under the influence of a voltage applied to the gate, electrons can tunnel into the nc-Al from the gate (or substrate) and tunnel out from the nc-Al towards the substrate (or the gate) depending on the polarity of the voltage. Electron tunneling can also take place between adjacent uncharged nanocrystals, and many such nanocrystals form conduction paths connecting the Si substrate to the metal gate as shown in Figure 5(a). For the virgin case, in addition to the one or two-dimensional transport, there may be some electron tunneling occurs in the third dimension, which makes $\zeta$ beyond $5/3$. The scenario of the influence of charge trapping in nc-Al on the tunneling paths is shown in Figure 5(b). Electron trapping in one nc-Al will affect the tunneling of other electrons into this nc-Al, and the relevant tunneling paths could be broken due to the Coulomb blockade effect. Therefore, as a result of the charging in nc-Al, with the breaking of some tunneling paths in the other two directions, the current conduction of the system will evolve towards the one-dimension transport. As the electric field of the applied voltage is along the vertical direction, the charges trapped in the paths along the vertical direction have a higher probability to tunnel out from the nanocrystals than those trapped in the paths along the lateral directions. Therefore, the charging in the paths along the lateral directions is more significant than in the paths along the vertical direction. The charging in the lateral directions causes the breaking of some tunneling paths in the lateral directions, leading to the evolution of the current conduction of the system towards the one-dimension transport.

4. CONCLUSIONS

In conclusion, Al-rich AlN thin films deposited by RF sputtering of Al target in an argon and N\textsubscript{2} gas mixture exhibit a memory effect. The memory effect is due to the charge trapping in the nc-Al embedded in the AlN matrix. The AlN thin films containing nc-Al provide the possibility of memory applications with low cost. On the other hand, the $I$--$V$ characteristic of the system follows a power law, i.e., $I = I_0 V^\zeta$. With charge trapping in nc-Al, $\zeta$ decreases towards $\zeta = 1$ of one-dimensional arrays, showing that the current conduction evolves with a trend towards the one-dimensional transport due to the breaking of some tunneling paths in the other two directions as a result of the charge trapping in nc-Al.

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References and Notes

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