Effects of target current density on the ionization rate of deposited particles and breakdown strength of AlN films

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Abstract
Aluminum nitride (AlN) films were deposited by DC reactive magnetron sputtering using two high purity aluminum targets, at selected target current densities (0.055, 0.083, 0.110 and 0.138 A cm\(^{-2}\)). The effects of the target current density on the ionization rate of Al atoms, morphology, chemical composition, crystal structure and breakdown strength of AlN films were studied by self-made device, SEM, XPS, XRD and withstanding voltage tester. It was found that the ionization rate of Al atoms gradually increased from 6% to 19% as target current density increased to 0.138 A cm\(^{-2}\). The results of XPS showed that the Al/N atomic ratio of AlN films gradually approached 1:1. Compared with low target current densities, the AlN films deposited at 0.110 and 0.138 A cm\(^{-2}\) exhibited a fine-crystal structure with average grain size <15 nm, good columnar structure with no obvious voids, and high breakdown strength. This indicated that Al atoms with high ionization rate could be applied to improve the insulation performance of AlN films.

1. Introduction
Aluminum nitride (AlN) films exhibit excellent properties such as, high thermal conductivity (320 W m\(^{-1}\) K\(^{-1}\)), high breakdown strength (14 MV cm\(^{-1}\)) and thermal expansion coefficient similar to Si [1–3]. These special properties of AlN films have great potential in optical, electronic and mechanical fields [4, 5]. Many methods can be used to prepare AlN films, namely molecular-beam epitaxy (MBE) [6], chemical vapor deposition (CVD) [7], pulsed laser deposition (PLD) [8] and DC reactive magnetron sputtering [9, 10]. Among these methods, DC reactive magnetron sputtering is a common method. As the morphology and chemical composition of AlN films could strongly influence the electrical, thermal and mechanical properties, it is necessary to understand the effects of deposition parameters of DC reactive magnetron sputtering on the structure and properties of AlN films.

Many research works have been done to studying the influence of deposition parameters the properties of AlN films. However, there are few studies on the influence of the ionization rate of Al atoms on the AlN films formation. In this work, AlN films with different ionization rates were deposited by DC reactive magnetron sputter. This work was devoted to study the effects of different ionization rates of Al atoms on the morphology and breakdown strength of the films. These works are expected to provide experimental and theoretical support for the research of insulating compound films.

2. Experimental section
Deposition of AlN films on P-type (100) Si and 6061 aluminum alloy substrates was carried out by DC reactive magnetron sputtering, in which two Al (Φ 120 mm × H 8 mm, 99.9%) targets placed oppositely. The substrates were placed at 80 mm from the Al targets, and the target discharge area was 36 cm\(^2\). The films deposition process lasted for 210 min at room temperature. The first 20 min was the etching and cleaning of Al targets and the
substrates in $2.5 \times 10^{-1}$ Pa with Argon atmosphere. Then Al base layer, AlN transition layer and AlN top layer were deposited sequentially. Specific deposition parameters are shown in table 1.

Ionization rate of Al atoms was detected by the self-made device, as shown in the figure 1. According to the classical Lorentz force theory, a uniform top-down magnetic field with a strength of 2000 GS was established in front of the Al target. Al particles (Al$^{++}$ and Al atoms) were sputtered out and entered the magnetic field in the deposition process. Al atoms did not deflect after entering the magnetic field, and were uniformly deposited on the left and right glass substrates. However, Al$^{++}$ were deposited on the left glass substrates. Thus the weight of Al films on the left glass substrates was heavier than that on the right glass substrates, and the weight difference was formed by Al$^{++}$. The percentage of the weight difference to the total weight of Al films on both sides was regarded as ionization rate of Al atoms. Among them, the Al films weight was measured by Sartorius-BSA224S electronic balance (Minimum weighing: 0.1 mg). The functional relationship between target current density ($I_d$) and ionization rate was as follows:

$$I = \frac{a - b}{a + b} \times 100\%$$ (1)

where $a$ is the Al films weight of the left glass substrates, and $b$ is the films weight of the right glass substrates.

The morphology and thickness of AlN films were observed from scanning electron microscope (SEM, JSM-6700F). The surface elements of the films were detected by x-ray photoelectron spectroscopy (ESCA-3600, voltage: 20 kV, current: 20 mA). The phase, grain size and crystallization rate of AlN films were analyzed by x-ray diffraction with CuKα anode (XRD-7000 s, voltage: 40 kV, current: 40 mA, scan speed: 10 deg min$^{-1}$), and the grain size of AlN films was estimated using Scherrer formula \([11-13]\). The breakdown voltage was measured by ZC71(D) withstanding voltage tester. To ensure the accuracy, the mean values of each sample was determined by 5 measurements.

### 3. Results and discussion

#### 3.1. Ionization rate of Al atoms

The measurement results on the Al films weight of the left and right glass substrates are shown in table 2. Before the experiment, a pre-test ($I_d: 0.055$ A cm$^{-2}$, no magnetic field) is implemented to prove that Al particles can be

| Processing          | Time (min) | $I_t$ (A) | $I_d$ (A cm$^{-2}$) | Ar/N$_2$ flow (sccm) | Substrate bias (V) |
|---------------------|------------|-----------|---------------------|----------------------|--------------------|
| Cleaning            | 20         | 0.3       | —                   | 10/0                 | —                  |
| Al base layer       | 5          | 0.3/2     | —                   | 10/0                 | —                  |
| AlN transition layer| 5          | 2         | —                   | 10/0/20              | —                  |
| AlN top layer       | 180        | 2/3/4/5   | 0.055/0.083/0.110/0.138 | 10/20              | —                  |

$I_t$—target current

$I_d$—target current density
evenly distributed on the left and right substrates without a magnetic field. The film weights of the left and right substrates are 0.0226 and 0.0228 g, respectively, with little difference. Ionization rate of Al atoms increases slightly with increasing \( I_d \) to 0.083 A cm\(^{-2}\), but the change is not obvious. As \( I_d \) continues to increase, the ionization rate of Al atoms increases qualitatively, up to 19%. In the traditional MSIP (\( I_d \) at 0.0001–0.1 A cm\(^{-2}\) [14]), formation of electrons and ions between the target and vacuum chamber mainly relies on collision ionization [15, 16]. The flux and initial energy of Al particles produced by bombarding the Al target increase to some extent with increasing \( I_d \) from 0.055 to 0.083 A cm\(^{-2}\), and the ionization rate of Al atoms increases slightly due to the increased probability of collision between Ar\(^+\) and Al atoms. At high \( I_d \) (> 0.083 A cm\(^{-2}\)), the temperature of the Al target surface quickly reaches the critical value of thermionic emission due to Ar\(^+\) bombardment and Joule-heating (\( Q = I^2 R_t \)) [17]. High-energy electrons escape from the Al target surface and drag out large-flux of Al atoms, leading to high ionization rate due to the collision of high-flux of electrons, Ar\(^+\) and Al atoms [14, 17].

3.2. Morphology and thickness of AlN films

Figure 2 shows the surface/section SEM images of AlN films using different \( I_d \). The AlN film at 0.055 A cm\(^{-2}\) exhibits spherical granules with different sizes and a loose porous columnar structure (figure 2(a) and (e)). At 0.083 A cm\(^{-2}\), the sizes of the spherical granules gradually increase to 0.5–0.6 \( \mu \)m and become uniform, and the number of micropores between the columnar crystals begins to decrease (figures 2(b) and (f)). The spherical granules sizes on \( I_d \) of 0.110 and 0.138 A cm\(^{-2}\) decrease rapidly, and there were no obvious micropores between columnar structures (figures 2(c), (g), (d) and (h)), indicating that the AlN films have good compactness. The morphology change could be attributed to the instantaneous high flux of Al particles and high \( I_d \). In addition, the average deposition rates of AlN films are 9.3, 10.3, 11.0 and 14.2 nm min\(^{-1}\) for different \( I_d \), respectively as shown in figure 3. In sputtering process, high energy and flux of Ar\(^+\) bombard the Al targets, causing the number of off-target Al particles to increase. Moreover, a sharp increase of ionization rate also means that thermionic emission phenomenon is more obvious that the escape of high-energy electrons from the Al target surface will drag out large-flux of Al atoms thus the average deposition rate increases [14, 18].

The above results confirm that when \( I_d \) is below 0.083 A cm\(^{-2}\), the Al atoms sputtered by Ar\(^+\) collision exhibit low ionization rate, low energy, and small-flux, leading to the granules mainly growing in an independent islands mode due to the low mobility. However, at \( I_d \) (> 0.083 A cm\(^{-2}\)), the Al atoms obtained by the collision and thermionic emission have the characteristics of high ionization rate, high energy and large-flux. The high-energy of Al particles exhibits high mobility and diffusivity on the substrate, which is conducive to migration to the micropores between the grains. On the other hand, under negative bias, the bombardment of high flux and energy of ions (Al\(^{n+}\) and Ar\(^+\)) can remove the protrusions on the films surface and the weaker structure, resulting in a flat surface quality and compact structure of the AlN films.

3.3. Composition of AlN films

Figure 4 exhibits the XPS full spectrum analysis of AlN films surface etched for 10 min at different \( I_d \). Al, N and O elements are detected in all AlN films. XPS is used to detect the Al/N atomic ratio of AlN films at different \( I_d \), as shown in figure 5. As \( I_d \) is 0.055 and 0.083 A cm\(^{-2}\), the Al/N atomic ratio is maintained above 1.13:1. However, the Al/N atomic ratio is maintained at about 1.06:1 on \( I_d \) of 0.110 and 0.138 A cm\(^{-2}\). This transition in atomic ratio could be attributed to the ionization rate of Al atoms. The Al particles are mostly low-energy neutral atoms due to low ionization rate (6% and 9%) of Al atoms, which is not conducive to prepare AlN films with Al/N atomic ratio of 1:1. Compared with low ionization rate, the proportion of high-energy Al\(^{n+}\) continues to increase, which promotes the combination of Al and N elements, and the Al/N atomic ratio of the prepared films is closer to 1:1.

### Table 2. Ionization rate of Al atoms at different \( I_d \) (working pressure: 2.8 \( \times \) 10\(^{-1}\) Pa, deposition time: 40 min, room temperature, target-substrate distance: 100 mm).

| \( I_d \) (A cm\(^{-2}\)) | Left Al films weight (g) | Right Al films weight (g) | Ionization rate (%) |
|-------------------------|--------------------------|---------------------------|---------------------|
| 0.055                   | 0.0244                   | 0.0216                    | 6                   |
| 0.083                   | 0.0285                   | 0.0238                    | 9                   |
| 0.110                   | 0.0383                   | 0.0283                    | 15                  |
| 0.138                   | 0.0488                   | 0.0332                    | 19                  |
3.4. Crystal structure of AlN films

Figure 6 displays the XRD patterns of AlN films at different $I_d$. The AlN film at 0.055 A cm$^{-2}$ exhibits a strong (101) peak. As $I_d$ increases, the intensity of (002) peak gradually increases. The change in crystallographic texture...
is due to the increase of the diffusivity of the particles, which makes it easier to diffuse to the lowest surface energy (002) position. Moreover, the decrease in (002) peak intensity at 0.110 Å cm$^{-2}$ is due to the bombardment of high-energy particles under the acceleration of negative bias, which destroys the growth of the (002) peak. The intensity of the (002) peak increases again, because the bombardment of high-energy particles lead to a temperature rise effect of the films, which in turn promotes the crystal growth of the films (shown in table 3) [19].

In the preparation process, the grain size and crystal structure of films can be affected by the transient flux and energy of deposited particles [20]. At low $I_d$ (<0.083 Å cm$^{-2}$), low energy and flux of Al particles in the plasma lead to low arrangement density and small grain size (shown in figures 2(a) and (b)) due to the low diffusivity and mobility of the particles. When $I_d$ is above 0.083 Å cm$^{-2}$, the Al atoms in the plasma are more
ionized and have more energy, but the faster deposition rate inhibits further growth of the film grains, limiting the film grain size to below 15 nm.

3.5. Breakdown strength of AlN films

The breakdown strength was used to evaluate the insulating properties of AlN films. The dependence of breakdown voltage and breakdown strength of AlN films at different $I_d$ is illustrated in Figure 7. When $I_d$ is 0.055 and 0.083 A cm$^{-2}$, the deposited films have lower breakdown strength ($< 148$ V $\mu$m$^{-1}$), while the films deposited at $I_d$ (0.110 and 0.138 A cm$^{-2}$) have greater breakdown strength. Among them, the 0.110 A cm$^{-2}$ film has the highest breakdown strength of 182 V $\mu$m$^{-1}$. This observation can be attributed to the improvement of the film arrangement density and Al/N atomic ratio. From the surface/section morphology of AlN films, during
the growth of the films, the low energy and ionization of Al atoms lead to poor breakdown strength due to loose structures and low Al/N atomic ratio. At 0.110 and 0.138 A cm$^{-2}$, high ionization and energy of Al atoms can not only overcome the influence of micropores to form a dense structure, but also promote the reaction between Al and N. However, the reduction of breakdown strength at 0.138 A cm$^{-2}$ may be attributed to the increase in crystallization and grain boundaries of AlN films, leading to increasing of the leakage current. This is consistent with Wang et al [21]’s research on breakdown strength.

4. Conclusion

In this study, AlN films with different ionization rates of Al atoms were deposited using DC reactive magnetron sputtering under various target current densities (0.055, 0.083, 0.110 and 0.138 A cm$^{-2}$). It was observed that the ionization rate of Al atoms increased gradually with increasing of $I_d$. Compared with low $I_d$, at high $I_d$ (>0.083 A cm$^{-2}$), the films formed by high energy and ionized Al atoms exhibited a dense fine crystal structure (<15 nm) and high breakdown strength (up to 182 V $\mu$m$^{-1}$). In short, during the deposition of AlN films, increasing ionization rate of Al atoms was conducive to improve the morphology and breakdown strength of AlN films.

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