Preparation of high-quality AlN films by two-step method of radio frequency magnetron sputtering*

ZHU Yu-qing (朱宇清), **CHEN Xi-ming** (陈希明)**, **LI Fu-long** (李福龙), **LI Xiao-wei** (李晓伟), **and YANG Bao-he** (杨保和)

School of Electronics Information Engineering, Tianjin University of Technology, Tianjin 300384, China

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The preparation of nanometer aluminum nitrogen (AlN) films with uniform lattice arrangement is of great significance for the manufacture of high-frequency surface acoustic wave (SAW) device. We put forward the two-step growth method and the annealing treatment method for the deposition of (100) AlN thin films. The results show that when the sputtering pressure is 1.2 Pa and the ratio between N_2 and Ar is 12:8, the influence of lattice thermal mismatch and anti-phase is the smallest during the nucleation growth at low-temperature stage of (100) AlN/(100) Si films. The root-mean-square (RMS) surface roughness of AlN prepared by the two-step method is reduced from 6.4 nm to 2.1 nm compared with that by common deposition process.

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With the development of high-frequency (above 5 GHz) and high-power surface acoustic wave (SAW) devices, aluminum nitrogen (AlN) thin films have been widely investigated[1] because of their high velocity and suitable piezoelectric coupling factors. In order to reduce the consumption of frequency and power during the propagation of SAW, the preparation of nanometer AlN films with uniform crystal lattice arrangement has become a research hot spot[2,3].

It is difficult to avoid the defects of films[4,5] during preparation by common radio frequency (RF) magnetron sputtering technique. Z. R. Songn[6] simulated the deposition process of AlN films, and the simulation results showed that the changes of lattice thermal mismatch and anti-phase have a great effect on the quality of crystal and micro-structure of (100) AlN/(100) Si films. Zou Wei-wei[7] used the Monte Carlo method to research the phenomenon of misfit dislocation during the deposition process of films, and the results showed that lattice thermal mismatch causes the changes of stress and anti-phase influences the arrangement of atoms in nanometer films. Guo Yan[8] prepared the films of (100) AlN by optimizing the conditions, and the results showed that the problem of lattice thermal mismatch has been improved, and the films of (100) AlN with low root-mean-square (RMS) surface roughness of 6.5 nm have been prepared. But the defect of atom arrangement is very serious.

In order to improve the problems of lattice thermal mismatch and anti-phase, this paper puts forward the two-step growth method and annealing treatment method for deposition of (100) AlN thin films, and the AlN films with preferred orientation of (100) are deposited on the substrate of n-type (100) Si. In this paper, the prepared films of (100) AlN have the low RMS surface roughness of 2.1 nm, and the arrangement of atoms in the films is uniform.

Because Si is a non-polar crystal, there is a problem about the establishment of polarity during the process of the deposition of AlN films on the (100) Si. Due to the different strengths of chemical bonds, the N atom is bonded to Si atom to form the Si-N bond, and then Al atom and N atom alternately grow. But there is a rare phenomenon that Al-Al bond and N-N bond may appear, because single atomic step may exist on the surfaces of Si, which leads to that the atoms grown on the both sides of step are dislocated. The kind of structural defect is anti-phase[9,10]. Because of the different coefficients of thermal expansion of AlN and Si, the anisotropic coefficient of thermal expansion of AlN makes the thermal mismatch of (100) AlN/(100) Si system more complicated[11].

In order to solve the problems, we put forward the two-step method and annealing treatment to grow the AlN films. The temperature of nucleation layer growth is lower than that of island growth. Due to the formation of nano-crystal on the surface, it can reduce the thermal mismatch of AlN, and at the same time, it changes the way of the formation of the dislocations[12]. By the method of annealing treatment, the internal structure of films is

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** E-mail：xmchen2006@126.com

changed and redistributed in order to avoid anti-phase and reduce the density of dislocations.

The AlN films were deposited on (100) Si substrate by RF magnetron sputtering device, and the sputtering target was pure Al (99.999%) with the diameter of 60 mm and the thickness of 5 mm. The substrate was tackled with ultrasonic cleaning by 5% HF in order to reduce SiO_2 on the surface of Si, and then the high-purity nitrogen and argon (99.999%) gases were introduced into the chamber.

In the first step of nucleation growth stage, the sputtering chamber was evacuated below 5×10^{-5} Pa, and the sputtering target was pre-sputtered for 15 min. Moreover, high-purity gases were introduced into the chamber, the sample rotation was opened, then the heating power was turned down in order to ensure the proper temperature, and at last the temperature was kept stable in a short period for nucleation growth. If the temperature is lower, the diffuse distance will be limited, and atoms will grow according to Volmer-Weber growth. On the contrast, if the temperature is higher, the diffuse distance will be longer, so they will grow according to Frank-van der Merwe growth [13,14] .

In the second step of deposition stage, in a nitrogen atmosphere, heating power was turned on, and when the temperature rising was stable, AlN thin films were prepared by the common method with the optimal deposition conditions. Finally, AlN films were annealed in nitrogen atmosphere.

In this paper, the crystalline and preferred orientation are studied by X-ray diffraction (XRD), and the RMS surface roughness is studied by atomic force microscope (AFM).

The parameters in Tab.1 show the different total sputtering pressures during nucleation growth at low temperature stage. Fig.1 shows the XRD patterns of the films at different total pressures with other parameters stable during nucleation growth stage. When the total pressure is 0.8 Pa, the crystalline of AlN films is poorer, and the AlN films rich in Al texture are deposited. With the increase of total pressure, the crystalline of (100) AlN films becomes better, and at the same time Al texture in the AlN films gradually disappears. When the total pressure is 1.2 Pa, the crystalline of (100) AlN film is the best. With the total pressure continues to increase, when the total pressure is 1.6 Pa, the crystalline of (100) AlN films becomes bad. The results show that on one hand, the pressure affects the concentration of the working gases and the amount of Al atoms bombarding the target, on the other hand, the level of the sputtering pressure affects the mean free path (MFP) of particles. By the gas dynamics theory, the MFP is approximately in inverse proportion to the pressure at a certain temperature. So during the low temperature nucleation growth stage of two steps, when the sputtering pressure is lower, AlN films rich in Al texture are deposited, because some of Al atoms have so large kinetic energy and there is not enough time for nitride to reach the substrate. When the sputtering pressure is higher, the MFP of sputtered particles decreases and the frequency of particle collision increases, so that it's bad for deposition of the AlN films, even that there are no films deposited on the substrate, and it leads to that the deposition effect of nucleation growth stage is not obvious. The problems of lattice thermal mismatch and anti-phase are not solved well during the deposition of AlN thin films, so that the crystalline of (100) AlN films becomes bad.

Tab.1 Parameters for the preparation of AlN thin films with different total sputtering pressures during nucleation growth at low temperature stage

Condition	Nucleation	Deposition
Total pressure (Pa)	0.8, 1.0, 1.2, 1.6	1.4
N_2:Ar	20:12	12:8
Power (W)	120	120
Al target-substrate distance (cm)	2.0	3.5
Substrate temperature (°C)	40	300
Time	12	120

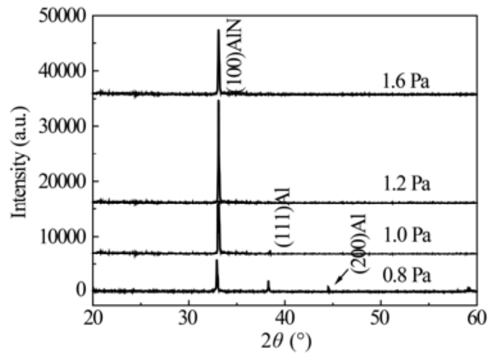
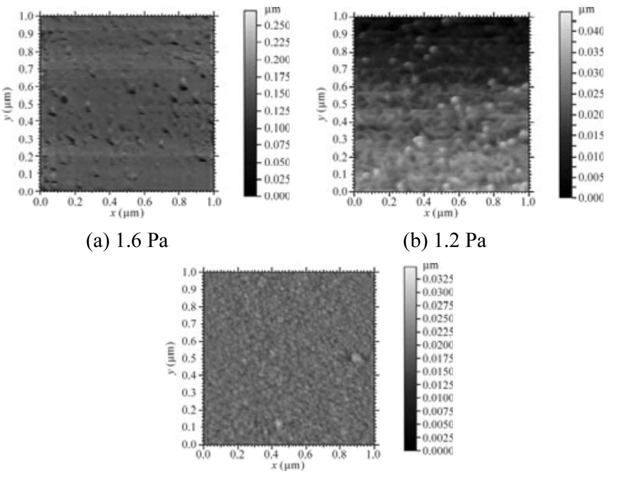


Fig.1 XRD patterns of AlN films at different total pressures

Fig.2 shows the AFM patterns of the AlN films at different total sputtering pressures with other parameters stable during nucleation growth stage. When the total sputtering pressure is 1.2 Pa, during nucleation growth, the RMS surface roughness of AlN films is 1.32 nm, and height difference of AlN films is 27.9 nm. AlN films show the obvious characteristics of nucleation growth, and the nucleation growth of films decides the chemical composition, the micro-structure and the defect of the films. As shown in Fig.1, in the common deposition of AlN films after the nucleation growth, the problems of stress and lattice thermal mismatch are improved, and the crystalline of (100) AlN films is the best. When the total sputtering pressure is 0.8 Pa, the RMS surface roughness of AlN films is 2.1 nm. Because the kinetic energy of Al is larger, there are many tiny Al atoms in the AlN films. When the total sputtering pressure is 1.6 Pa, the height difference of AlN films is 10.5 nm, and AlN films show obscure characteristics in nucleation growth.

Tab.2 shows the parameters for the preparation of AlN films with different ratios between N_2 and Ar during nucleation growth at low temperature stage. Fig.3 shows XRD patterns of the films with different ratios between N_2 and Ar and other parameters stable during nucleation growth stage. When ratio between N_2 and Ar is 18:14, the crystalline of (100) AlN film is the worst and the AlN films rich in Al texture are deposited. With the increase of ratio between N_2 and Ar, the crystalline of (100) AlN film becomes better. When ratio between N_2 and Ar is 22:10, the crystalline of (100) AlN film is the best. With the ratio of N_2 and Ar continues to increase, the crystalline of (100) AlN film becomes bad. The results show that N_2 participates in both sputtering and reaction during the process of nucleation growth at low temperature, at the same time, the kinetic energy of Ar^+ is larger than that of N^+, so sputtering yield of N_2 is less than Ar. When the ratio between N_2 and Ar is lower, although more Al atoms which have large kinetic energy are bombarded down from the substrate by the sputtering atoms, there is not enough N_2 which participates in reaction. When the ratio between N_2 and Ar is higher, it can lead to the decrease of the number of Al atoms, which are bombarded down from the substrate, and their kinetic energy, so that the number of Al atoms is small, even that there is no film deposited on the substrate. The problems of lattice thermal mismatch and anti-phase are not solved during the deposition of AlN thin films.



(a) 1.6 Pa (b) 1.2 Pa (c) 0.8 Pa

Fig.2 AFM patterns of AlN films at different total pressures during nucleation growth at low-temperature stage

Tab.2 Parameters for preparation of AlN thin films with different ratios of N_2 to Ar during nucleation stage

Condition	Nucleation	Deposition
Total pressure (Pa)	1.2	1.4
N_2:Ar	18:14, 20:12, 22:10, 24:8	12:8
Power (W)	120	120
Al target-substrate distance (cm)	2.0	3.5
Substrate temperature (°C)	40	300
Time	12	120

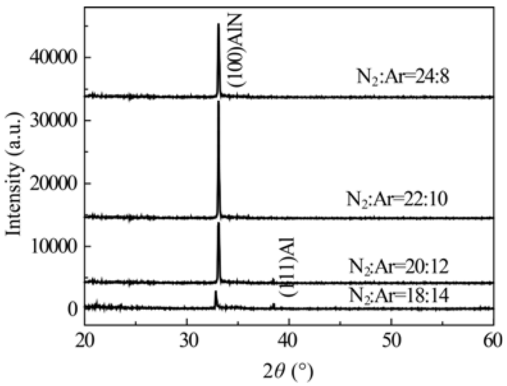
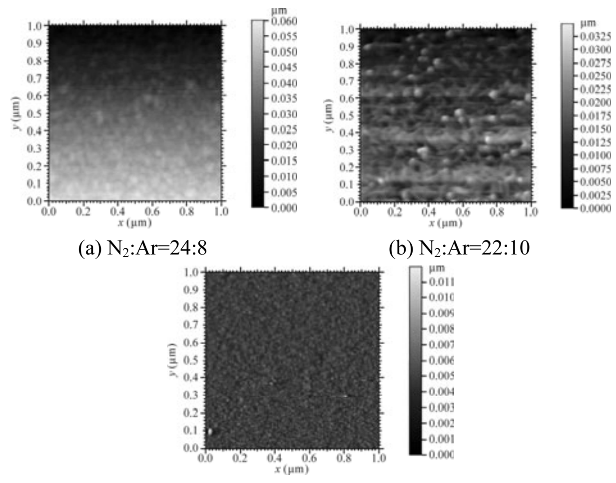


Fig.3 XRD patterns of AlN films at different ratios of N_2 to Ar

Fig.4 shows the AFM patterns of the AlN films with different ratios between N_2 and Ar and other parameters stable during nucleation growth stage. When the ratio between N_2 and Ar is 22:10, during nucleation growth, the RMS surface roughness of AlN films is 1.45 nm, the height difference of AlN films is 38.9 nm, and AlN films show obvious characteristics of nucleation growth. As shown in Fig.3, after the nucleation growth, the problems of dislocation stress and lattice mismatch are improved, and the crystalline of (100) AlN films is the best. When the ratio between N_2 and Ar is 18:16, there are many tiny Al atoms in the AlN films. When the ratio between N_2 and Ar is 24:8 which is lower, during nucleation growth, the RMS surface roughness of AlN films is 0.8 nm, and AlN films show obscure characteristics of nucleation growth.

Fig.5 shows the XRD patterns of the films by common method and the two-step method. The results show that the crystalline of AlN thin films deposited by the two-step method is much better than that by the common method. The crystalline of the AlN films deposited by the common method is not good because of lattice mismatch. At the same time, there are Al atoms in AlN films.



(a) N_2:Ar=24:8 (b) N_2:Ar=22:10 (c) N_2:Ar=18:16

Fig.4 AFM patterns of AlN films at different ratios of N_2 to Ar during nucleation growth at low temperature stage

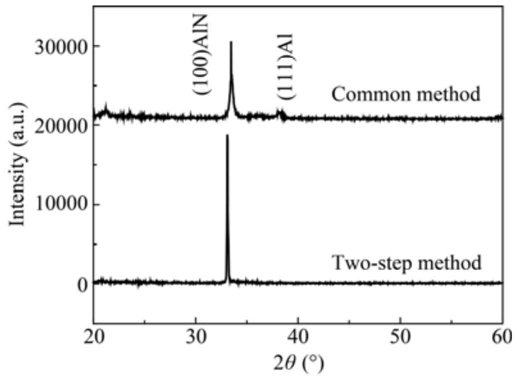
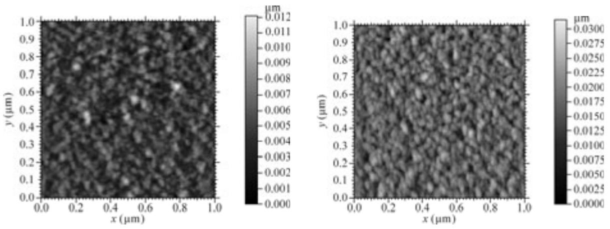


Fig.5 XRD patterns of AlN films deposited by two-step method and common method

Fig.6 shows the AFM patterns of the AlN films deposited by common process and two-step method, and the scanning range is 1 μm×1 μm. The RMS surface roughness of AlN films deposited by the common method is 6.4 nm, and the atom arrangement of film surface is more sparse. On the contrast, the RMS surface roughness of AlN films deposited by the two-step method is 2.1 nm. At the same time, the atom arrangement of film surface is more compact, and the surface of AlN films is uniform and flat.



(a) Common method (b) Two-step method

Fig.6 AFM patterns of AlN films deposited by two-step method and common method

In this paper, AlN films are deposited with annealing treatment method and two-step growth method by RF magnetron sputtering system. In the two-step growth method, the first step is the nucleation growth at low temperature, and then films are deposited by common method. This way improves the crystalline and micro-structure of AlN films and the stress caused by lattice thermal mismatch during the deposition. At the same time, this way solves the problem of anti-phase. At last, the RMS surface roughness of the (100) AlN/(100) Si films deposited by two-step method is 2.1 nm, and the arrangement of atoms is uniform.

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