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Preparation of AlN thin films for film bulk acoustic resonator application by radio frequency sputtering

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Abstract: Aluminum nitride (AlN) thin films with high *c*-axis orientation have been prepared on a glass substrate with an Al bottom electrode by radio frequency (RF) reactive magnetron sputtering. Based on the analysis of Berg's hysteresis model, the improved sputtering system is realized without a hysteresis effect. A new control method for rapidly depositing highly *c*-axis oriented AlN thin films is proposed. The N_2 concentration could be controlled by observing the changes in cathode voltage, to realize the optimum processing condition where the target could be fixed stably in the transition region, and both stoichiometric film composition and a high deposition rate could be obtained. Under a 500 W RF power of a target with a 6 cm diameter, a substrate temperature of 450 °C, a target-substrate distance of 60 mm and a N_2 concentration of 25%, AlN thin film with preferential (002) orientation was deposited at 2.3 μm/h which is a much higher rate than previously achieved. Through X-ray diffraction (XRD) analysis, the full width at half maximum (FWHM) of AlN (002) was shown to be about 0.28°, which shows the good crystallinity and crystal orientation of AlN thin film. With other parameters held constant, any increase or decrease in N_2 concentration results in an increase in the FWHM of AlN.

Key words: Aluminum nitride (AlN), Piezoelectric thin film, Radio frequency (RF) reactive sputtering, Preferred orientation, Film bulk acoustic resonator (FBAR)

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INTRODUCTION

Thin film bulk acoustic resonator (FBAR) has become one of the most promising components for the realization of microwave monolithic integrated circuits (MMICs), especially in high-frequency passive devices applicable to GHz-band wireless communication systems (Ruby *et al*., 1999; Tai *et al*., 2004). Conventionally, microwave ceramic resonators and surface acoustic wave (SAW) resonators have been applied in this frequency range. However, microwave ceramic resonators tend to be physically bulky, while SAW resonators have large insertion losses as well as limited power handling characteristics. FBAR devices can replace conventional SAW resonators and microwave ceramic resonators, since they offer several prominent advantages such as small size, high quality factor, high power handling capability, high operating frequency, good temperature stability, and the possibility of being integrated into radio frequency integrated circuit (RFIC) (Lakin, 2003; Weigel and Rappel, 2005). The most critical factor that determines the characteristics of FBAR is the material property of piezoelectric thin films, which is considered to be directly related to the preferred orientation of the crystal structure of deposited films.

The main methods for growing AlN thin films consist of chemical vapor deposition (CVD) techniques (Yang *et al*., 2003), molecular beam epitaxy (MBE) (Mansurov *et al*., 2007), laser ablation (Ristoscu *et al*., 2005), and direct current (DC) or RF reactive magnetron sputtering techniques (Yu *et al*., 2005; Medjani *et al*., 2006). Among all the methods,

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RF reactive magnetron sputtering is considered to be the optimal method so far but it can lead to a hysteresis effect (Berg and Nyberg, 2005); that is, in the flow control reactive sputtering system, more than one reactive state might exist corresponding to one particular N_2 pressure. Once the transition region is passed, the initial state cannot be recovered by changing only processing parameters, which is a disadvantage for fabricating AlN thin films industrially (Sproul *et al*., 2005). To obtain high deposition rates as well as optimum film properties, it is desirable to operate in the transition region between the metallic and poisoned states of the target. However, with flow control of the reactive gas, the process is unstable in the transition region in most cases. This problem leads to variability in AlN fabrication, which is one of the reasons for disagreement among researchers. Published results are sometimes conflicting and inconsistent (Okano *et al*., 1992; Huang *et al*., 2005; Chiu *et al*., 2007). Therefore, the avoidance of the hysteresis effect and the control of the reactive process are the key issues in the AlN fabrication process.

There are many factors that influence the growth of AlN films including sputtering power, gas pressure, target-substrate distance, N_2 concentration, and substrate temperature (Okano *et al*., 1992). In this work, we focus on the correlation between the N_2 concentration and the crystal orientation of films. Based on analysis of the Berg's model of reactive sputtering, we present a new method for achieving a high deposition rate as well as high *c*-axis oriented AlN thin films. In the improved reactive sputtering system, the hysteresis effect could be eliminated and the process could be stably controlled in the transition region. We also provide a new way of determining the quantity of N_2 flow by surveying the changes in cathode voltage.

ANALYSIS OF THE HYSTERESIS EFFECT OF REACTIVE SPUTTERING

Reasons for the hysteresis effect

A typical process curve showing the hysteresis effect is presented in Fig.1, which shows a plot of the cathode voltage vs the flow of reactive gas, divided into three different regions. In the metal mode region of the AlN thin film deposition process, due to the low partial pressure of the reactive gas N_2, the target is only marginally covered with compound and the sputtering process is almost identical to that of the pure metal Al target characterized mainly by high sputtering yield.

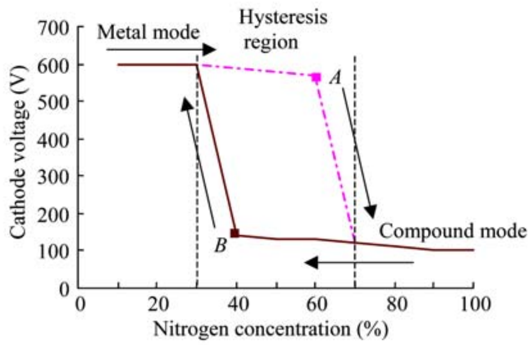


Fig.1 Typical hysteresis effect in reactive sputtering

With the slow increase in N_2 flow, the target surface becomes partially reactive with N_2 resulting in a slight decrease in the deposition rate, and the process transfers to the transition region (hysteresis region) where a tiny increase or decrease of reactive gas could greatly influence the process behavior. When the flow of N_2 is initially increased, its partial pressure in the chamber and the coverage of the target surface with nitride increase, so the fraction of exposed Al on the target decreases. This leads to a reduction in the consumption rate of N_2. However, this results in a further increase in the partial pressure of N_2, which in turn causes an even larger fraction of the target surface to be nitrided and so on. At this point (Point *A*), the deposition state changes uncontrollably until the whole target surface is poisoned as a result of a steep drop in the deposition rate, a steep jump in the N_2 partial pressure as well as the complete nitridation of the target. Meanwhile, the cathode voltage of the target also decreases steeply since the secondary electron emission coefficient of compounds is usually higher than that of pure metal surfaces.

When the entire target surface is covered with the compound AlN, it reaches compound mode, also named poisoned mode. In this mode, any further increase in the N_2 partial pressure does not affect the process significantly and the deposition rate drops almost over one order of magnitude compared to the elemental deposition rate for the same amount of power delivered to the target.

If at this moment the nitrogen flow is decreased, the transition region and metal mode will not occur at the same N_2 flow point (Point *A*) because the N_2 partial pressure is rather high compared with that when the target was operated in metal mode. The only way to decrease this high N_2 partial pressure to its original point is to further decrease the N_2 flow until the second transition point (Point *B*) is reached. So the deposition state steeply changes from compound mode to metal mode.

We suppose that if the hysteresis effect could be eliminated in the reactive sputtering process, the cathode voltage of the target would be the continuous and monotone function of the reactive gas flow, and the transition region would be distributed smoothly and gradually between the metal mode and the compound mode, as in Fig.2. To obtain high deposition rates and optimum film properties, it is desirable to operate in the transition region between the metal mode and the compound mode, for example, at Point *C*. However, in most cases with flow control of the reactive gas, the process is unstable in the transition region which leads to uncertainty in AlN fabrication (Sproul *et al*., 2005).

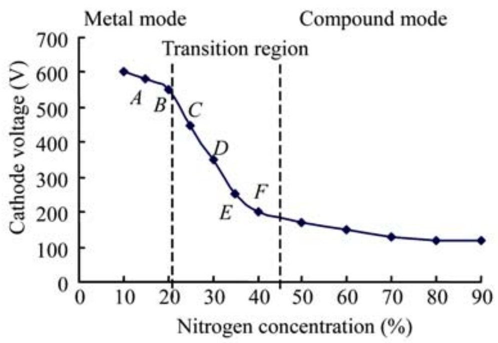


Fig.2 Improved sputtering system (without hysteresis effect)

Employing Berg's model to analyze AlN fabrication process

To analyze how the hysteresis is affected by different processing parameters, we employ Berg's model (Berg and Nyberg, 2005). In the mathematical model that describes the reactive sputtering system, all the parameters that influence the hysteresis effect are defined and simulated. According to his theory, there exist conditions under which hysteresis does not occur. He presented a useful equation displayed as $\frac{dQ_{tot}}{dP}=\frac{dQ_c}{dP}+\frac{dQ_t}{dP}+\frac{dQ_p}{dP}$, which could determine whether the hysteresis will appear or not, where Q_{tot} represents the total supply rate of the reactive gas, Q_c the gas consumption at the substrate, Q_t the gas consumption at the target, Q_p the gas to pump and $\frac{dQ_p}{dP}=S$ denotes the pumping speed of the system. If $\frac{dQ_{tot}}{dP}<0$ in some region, the process will exhibit hysteresis; while if $\frac{dQ_{tot}}{dP}>0$ over the whole processing region, the process will not exhibit any hysteresis.

Fig.3a displays the simulation results about the calculated sputter erosion rates, *R*, vs reactive gas supply for three different pumping speeds, *S*. It shows that there will be no hysteresis at high pumping speeds. Fig.3b shows the calculated sputter erosion rates, *R*, vs reactive gas supply for different target areas, A_t. The simulations indicate that the hysteresis may be eliminated if the target area is small. All these models predict that the shape of the processing curves depends strongly on the parameters of the sputtering process. Simulations involving all the parameters in the sputtering process indicate that hysteresis could be avoided by reducing the target-substrate distance, reducing the target area, increasing the system pumping speed, and so on. Consequently, the optimum processing conditions would be realized where the target could be fixed stably in the transition region and both stoichiometric film composition and a high deposition rate could be obtained.

EXPERIMENTAL DETAILS

The AlN films were prepared on a glass substrate with an Al bottom electrode using the reactive RF magnetron sputtering technique. We improved the experimental system according to the Berg's theory to eliminate the hysteresis effect. The pumping speed was chosen to be 1200 L/s, the target-substrate distance was fixed at 60 mm and the size of the target was 6 cm. The purities of Ar and N_2 were 99.999%. The glass substrate was first etched in a concentration of 0.5% (v/v) hydrogen fluoride solution for 10 min to remove surface impurity, followed by oven-baking for 30 min at 100 °C. Subsequently, a 200 nm aluminum film was deposited in an Ar atmosphere

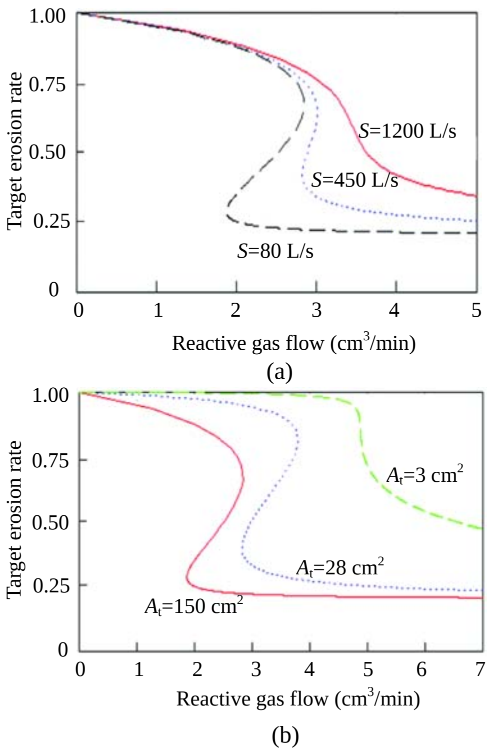


Fig.3 Calculated sputter erosion rates, *R*, vs reactive gas supply (a) for three different pumping speeds, S; (b) for different values of target areas, A_t

by DC sputter to form the bottom electrode. Then an AlN thin film was deposited on top of the aluminum layer in an Ar/N_2 reactive atmosphere in the desired sputtering conditions. During the deposition process, an impedance matching network was used to control the RF power input. The base pressure and sputtering pressure were chosen to be 3×10^{-4} and 0.23 Pa, respectively. The substrate was heated directly at 450 °C. Several sputtering runs were carried out at various N_2 under the control of N_2 flow to evaluate the optimum deposition conditions. Details of the deposition conditions are shown in Table 1.

The crystalline structure and crystal orientation of each AlN film were characterized by X-ray diffraction (XRD) analysis with X'Pert PRO (PANalytical) using Cu K_a radiation and scanned from 2θ 20°~60°. The degree of *c*-axis crystallization was examined from the full-width at the half maximum (FWHM) of the AlN (002)-diffraction peak. For surface morphology and cross-sectional columnar structure observation, samples were cut and the microscopic film was observed using a Scanning Electron Microscope (SEM) (SLR10N, FEI Company, the Netherland).

RESULTS AND DISCUSSION

The XRD patterns of a set of AlN thin films deposited on glass substrates with Al electrodes in different N_2 concentration are shown in Fig.4. The XRD peaks of AlN films are observed near 2θ=36.1°, which corresponds to the AlN (002) reflection. These patterns indicate that the AlN thin films were perpendicular to the substrate surface. The Al (111) planes could also be observed near 2θ=39° just next to the AlN (002) planes. Fig.5 presents the peak intensity counts of AlN crystallographic variation dependent on the N_2 concentration.

Passing from the sample deposited at a N_2 concentration of 15% to the one deposited at a N_2 concentration of 40%, an initial increase and then a decrease of the AlN (002) planes were found. The intensities of AlN films did not increase or decrease simply with an increase in N_2 concentration. Instead, there was a certain N_2 concentration at which the intensity of AlN films reached an optimum value. At a N_2 concentration of 15%, a preferred orientation

Table 1 Sputtering conditions of Al and AlN films

Parameter	Film material	
	Al	AlN
Process	DC magnetron sputtering	RF reactive magnetron sputtering
Target	Al 99.999% purity, 6 cm diameter	Al 99.999% purity, 6 cm diameter
Power (W)	100	500
Substrate temperature (°C)	Room temperature	450
Target substrate distance (mm)	60	60
Sputtering pressure (Pa)	0.38	0.23
Deposition time (min)	2	60
Atmosphere	Ar	$n_{N_2}:n_{N_2+Ar}$=15%, 20%, 25%, 30%, 35%, 40%
Cathode voltage (V)	–	580, 550, 450, 350, 250, 200

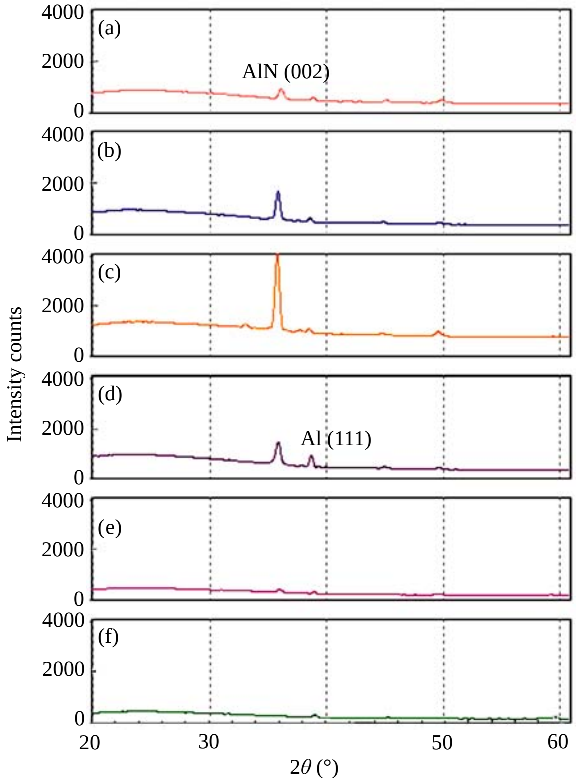


Fig.4 X-ray diffraction patterns of AlN thin films deposited on glass substrates with Al electrodes under different N_2 concentrations. (a) 15%; (b) 20%; (c) 25%; (d) 30%; (e) 35%; (f) 40%

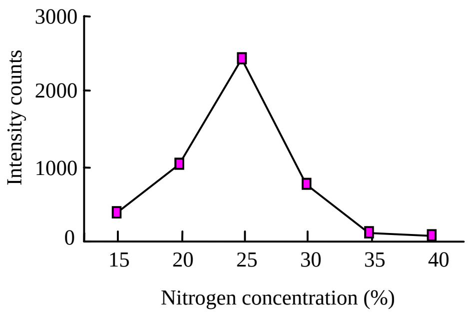


Fig.5 Dependence of peak intensity of AlN (002) planes on different N_2 concentration

along (002) direction at 2θ=36.1° was noticed, but the intensity was not distinct; with the N_2 concentration increased to 20%, the intensity of *c*-axis orientation seemed to be stronger; at a N_2 concentration of 25%, the film was oriented along the *c*-axis in a preferred direction with quite strong intensity. As the flow of N_2 was further increased, however, the intensity of AlN (002) orientation at 2θ=36.1° began to decrease. At a N_2 concentration of 30%, the contributions of the two domains, including another Al (111) orientation, were comparable and the peak intensity of AlN *c*-axis orientation decreased drastically compared with that at 25% N_2 concentration; at a N_2 concentration of 35%, the AlN (002) plane peak was even weaker and at the N_2 concentration of 40%, it was almost unnoticeable.

Fig.6 is a plot of the dependence of the full width at half maximum (FWHM) of AlN (002) orientation against the N_2 concentration. It shows that the FWHM degree had a minimum of 0.28° at a N_2 concentration of 25%. Fig.7 is a plot of the dependence of the deposition rates on the N_2 concentration indicating that the deposition rate fell as the N_2 concentration increased.

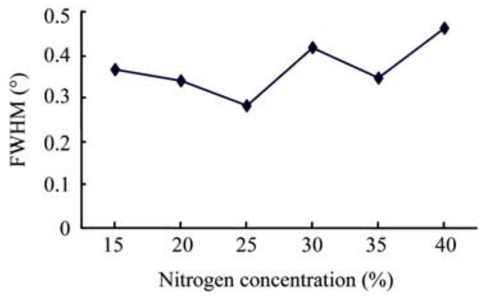


Fig.6 Dependence of FWHM degree of AlN (002) planes on different N_2 concentration

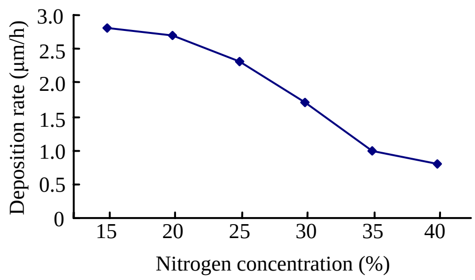
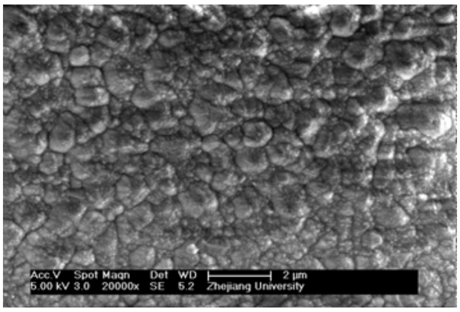


Fig.7 Dependence of AlN film deposition rate on different N_2 concentration

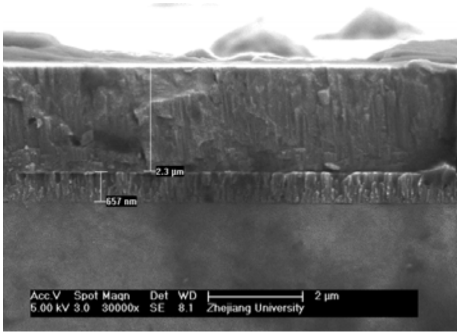
All these phenomena can be explained completely by Berg's model. Experimental results showed that a reactive sputtering system without hysteresis can be achieved under our desired conditions (Fig.2). As the N_2 flow slowly increased, the cathode voltage dropped from 600 to 150 V and the reverse process could be obtained as the N_2 flow was decreased. That is, there is only one N_2 flow point corresponding to one particular cathode voltage and the cathode voltage changes as the N_2 flow varies, resulting in different working states of the target. Consequently, the sputtering process can be controlled at the transition region, where a high deposition rate as well as stoichiometric film composition is guaranteed, by adjusting the N_2 flow. The maximum and minimum of the cathode voltages were 600 and 150 V, corresponding to the metallic mode of the target with 0% N_2 concentration and the compound mode of the target with 100% N_2 concentration, respectively. Points *A*, *B*, *C*, *D*, *E* and *F* at which the cathode voltages are 580, 550, 450, 350, 250 and 200 V with a N_2 concentration of 15%, 20%, 25%, 30%, 35% and 40%, respectively, represent three different target modes under which the samples were fabricated. It is obvious that the optimal N_2 concentration is that when the cathode voltage is fixed around the mid-point between the maximum and the minimum, and in our work the optimum value of N_2 concentration was 25%.

We suggest that as the N_2 concentration was increased until it reached the mid-point of the cathode voltage (from 15% to 25%), the sputtering process changed gradually from the metallic mode into the transition mode and in the process more sputtered Al ions would have been nitrified before they reached the substrate. Although the deposition rate slowly decreased, the stoichiometry of the film was improved, leading to a distinct favoring of the *c*-axis orientation. However, when the N_2 concentration was further increased to 40%, the target surface was covered with the compound AlN layer since more nitride compounds were formed at the target surface before the Al atoms were sputtered onto the substrate. Therefore, the deposition rate further decreased and the sputtering process changed to the compound mode. Since the Ar flow was never changed, increasing the N_2 concentration meant increasing the sputtering pressure. Thus, the Al and N atoms would have had to collide with more other atoms or ions before arriving at the substrate surface, and would not have had enough energy to arrange the texture according to the hexagonal wurtzite crystalline structure at the substrate surface. Accordingly, the intensity of the preferred orientation was weakened and the degree of FWHM was affected.

Fig.8a shows a top view SEM photograph of AlN film prepared under the optimum conditions with a N_2 concentration of 25% in this work, displaying the dense and smooth surface features of AlN film. Fig.8b is a SEM micrograph showing the cross-sectional structure of AlN film lying on an Al electrode and a glass substrate, indicating that the AlN film grown on the Al electrode exhibits a typical oriented columnar structure and has a fine-grain structure in the vicinity of the Al surface. This improves the coupling factor when these films are used in FBAR devices.



(a)



(b)

Fig.8 SEM micrographs of AlN film deposited on an Al bottom electrode at a substrate temperature of 450 °C, RF power of 500 W, working pressure of 0.23 Pa, target-substrate distance of 60 mm, N_2 concentration of 25%. (a) Surface morphology of AlN film; (b) Cross-sectional SEM of AlN film

CONCLUSION

A hysteresis effect exists in reactive sputtering systems which can lead to uncertainty of processing control. In this study, employing Berg's model on the reactive sputtering processes, we provide a new method for achieving a high deposition rate as well as high *c*-axis orientation for AlN thin films. The proposed method uses the flow of N_2 as a control quantity, and the cathode voltage as a location quantity. The optimal N_2 concentration is that found when the cathode voltage is located at the mid-point between the maximum and minimum; in our study that value was 25%. The experimental results revealed that AlN films prepared on glass substrate with Al electrodes using this method have enhanced *c*-axis orientation and exhibit highly textured morphologies. The deposition rate reached 2.3 μm/h at an RF power of 500 W, target-substrate distance of 60 mm and substrate temperature of 450 °C, which is of great importance in the industrial fabrication process.

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