Influence of technological regimes of plasma chemical deposition on texture formation in aluminum nitride films

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Abstract. Textured polycrystalline aluminum nitride films were grown by magnetron sputtering of an aluminum target in a mixture of argon and nitrogen, followed by the formation of a film on sitall substrates.  The influence of the pressure of the working gases, the power dissipated on the target, and the ratio of the Ar / N₂ gas fluxes on the texture of the aluminum nitride films are investigated. The phase composition and degree of crystallinity of the obtained films were determined using scanning electron microscopy and X-ray diffraction.  The results of studies of the phase composition and parameters of the crystal structure of aluminum nitride films are presented: the size of the coherent scattering regions, the predominant orientation of the crystallites in the growing film, and the degree of crystallinity. It is noted that the films are predominantly oriented along the (111) and (002) planes. The degree of crystallinity of the A1N films varied from 10 to 30% depending on the conditions of the deposition process. The relative microdeformation of the lattice (Δd/d), depending on the parameters, ranged from 1.7·10⁻³ to 6.9·10⁻³. The size of the crystallites is affected by the microdeformation of the lattice, with its increase by 4 times, the size of the crystallites decreased by 3.6 times from 29 to 8 nm.

1. Introduction
Among semiconductor materials, aluminum nitride occupies a special place due to the combination of such technological parameters as high thermal conductivity, the value of the constant dielectric constant, temperature stability, electromechanical coupling coefficient, and the speed of sound propagation. The above properties, as well as the low coefficient of thermal expansion at high temperatures, determine the prospects for the use of aluminum nitride films in acoustoelectronic devices, in particular, in devices based on surface acoustic waves (PAW). PAW resonators are widely used in various sensors of physical quantities, filters and generators.

The determining role in the design and manufacture of devices for selecting and generating signals based on thin-film technologies is played by the technological parameters that provide the target properties of the structural materials of films as resonator elements (control of the mechanism of texture formation in films, the degree of crystallinity, the predominant orientation of crystallites in the film, and coherent scattering regions (OCG)).

In the process of forming aluminum nitride films by magnetron sputtering, the texture of the predominant orientation of the crystallites is formed. The practical interest in film textures is related to the fact that they cause anisotropy of the properties of the matrix crystallites along the acoustic wave...
propagation vectors. The problem of modeling the mechanisms of growth and texture formation of aluminum nitride films used in acoustoelectronics is becoming more and more relevant. Many works are devoted to the study of the textures of aluminum nitride films since the texture is associated with the properties of this material that affect the operation of acoustoelectric devices based on them [7-10].

The crystal properties of the structure of aluminum nitride films were studied by X-ray diffraction on a DRON-4 diffractometer (CuKα radiation) [11-12].

The work aims to determine the dependence of the crystallographic characteristics of the aluminum nitride film material and the surface morphology on the technological parameters of the deposition conditions.

The paper presents the results of a study of the crystallographic properties and surface morphology of aluminum nitride films.

2. **Research objective**

To simplify and reduce the cost of the production technology of acousto-electronic devices based on multilayer structures, sitall substrates are used. When using such non-monocrystalline non-textured substrates, the problem arises of obtaining continuous homogeneous films with a certain texture.

Aluminum nitride films have spontaneous polarization and exhibit a piezoelectric effect in the direction of the orientation of the crystallites of the structure <001>, along the polar C axis, so the formation of AlN films with the orientation of the C axis perpendicular to the substrate surface is of interest.

It is known that the texture of films is greatly influenced by the substrate temperature, which determines the mechanism of condensation and growth of films, as well as the quality of the substrate surface, since nucleation occurs on surface defects [11].

The problem of determining the influence of other technological parameters of plasma-chemical magnetron sputtering in the formation of multilayer Al-AlN films with a given structure, composition, and morphology on the phase composition and structural parameters of samples for functional electronics devices remains relevant. The correlations of phase and structural transformations, structure, mechanical stresses, and physical properties of layers of AlN-based multilayer films remain insufficiently studied at present.

The main tasks of this work are:

- determination of the parameters of the crystal structure (size, shape, and orientation of crystallites) of aluminum nitride films;
- determination of the relationship between the characteristics of films and the processes and technological modes of magnetron formation of targeted films;
- determination of the most optimal modes of formation of multilayer structures based on AlN films.

3. **Experimental technique**

Aluminum nitride films were formed by magnetron sputtering of an aluminum target (99.999%) at direct current in a nitrogen and argon atmosphere on a vacuum multi-component coating plant STEMS116-01. Argon consumption varied from 6 to 8 sccm, nitrogen consumption was 8 sccm. The pressure in the chamber varied from 0.065 to 0.9 Mpa, the power on the target from 850 to 950 watts. The aluminum target was attached to a water-cooled cathode. The vacuum chamber was previously pumped out by a turbomolecular pump to a pressure of no more than 2·10^{-4}Pa. Previously, the substrate was heated by IR radiation up to a temperature 350K. Sitall substrates of the ST50 brand-1-1-0.6 were located above the target at a distance of 100 mm. The thickness of the aluminum nitride film was measured using the MII-4 micro interferometer.

The effects of the gas mixture pressure, the DC magnetron discharge power scattered on an aluminum target, the concentration of argon and nitrogen on the parameters that determine the crystal properties of the film: lattice parameters, crystallite sizes, the degree of crystallinity, lattice deformations, and the orientation of the texture axis were studied.
The phase composition and parameters of the crystal lattice were studied using a DRON-4 diffractometer based on CuKα radiation. The data was analyzed by the POWDER CELL 2.4 program. The research was carried out in the Materials Science Center for Collective Use of TSU, Tomsk.

4. Experimental results and discussion

The quantitative and qualitative phase analysis was carried out, the parameters of the unit cell, the type of lattice, and the spatial group were determined. Quantitative analysis made it possible to determine the content of crystal phases in the films, the size of the coherent-scattering regions, the values of the relative lattice micro deformation ($\Delta d/d$), and the degree of film crystallinity. The data of the technological parameters of sputtering and the studied parameters of the samples formed by the films are given in Table 1.

Aluminum nitride films with a thickness of 0.5 microns were studied on sitall substrates, at the same substrate temperature and different working gas pressures and capacities on the target. The size of the coherent scattering region was taken as the size of the crystallites.

Table 1. Technological parameters of sputtering, parameters of the study of the formed films.

| Sample no | 1 (75) | 2 (77) | 3 (78) | 4 (85) |
|-----------|--------|--------|--------|--------|
| Ar flow, sccm | 8 | 6 | 8 | 7 |
| $N_2$, flow sccm | 8 | 8 | 8 | 8 |
| Pressure, Pa | 0.09 | 0.065 | 0.08 | 0.07 |
| Power on the target, W | 850 | 850 | 950 | 900 |
| Detected phases | AIN 186 hex orient 10% along (002) | AIN 216 CUBIC advantage. orient. 76 % along (111) | AIN 186 hex orient 45 % along (002) | AIN 216 CUBIC, advantage. orient. 69 % along (111) |
| The content of the phases, mass% | <5 | 21 | 35 | 9 |
| The lattice parameters, Å | a = 3.1149 c = 5.0577 | a = 4.4055 | a = 3.1660 c = 4.9157 | a = 4.3403 |
| CSR size, nm | 29 | 14 | 8 | 21.3 |
| $\Delta d/d \times 10^{-3}$ | 1.7 | 2.14 | 6.9 | 2.14 |
| Crystallinity degree, % | 25-30 | 10-15 | 25-30 | 25-30 |

Depending on the technological parameters of the deposition process, the degree of crystallinity of the films formed by this method varies from 10 to 30%.

The value of the relative microstrain of the lattice ($\Delta d/d$) depending on the parameters, ranged from $1.7 \times 10^{-3}$ to $6.9 \times 10^{-3}$, with an increase in microstrain, the CSR size decreased.

Thus, the results of X-ray diffractometry showed that the A1N films No. 75 and No. 85 are strongly textured in the direction (111), and the films No. 77 and No. 78 in the direction (002).
The detected phases with a predominant orientation (002) in the crystal lattice of AlN films indicate that the aluminum nitride layers have a single-crystal hexagonal structure perpendicular to the substrate surface (Fig. 1).

According to X-ray diffractometry, an intense reflection from the film surface is observed at an angle of 2θ=36°. According to the table values, it is an aluminum nitride with a hexagonal structure with an axis directed perpendicular to the surface of the growing film.

The surfaces of the films with both orientation (111) and orientation (002) have a textured structure with columnar crystallites that persist along the entire thickness of the film.

Additional studies of the surface morphology of the grown films using a Hitachi SU8280 scanning electron microscope also revealed a columnar structure.

Figure 2 shows a snapshot of the morphology of the film surface.

Figure 1. X-ray swing curve of the AlN layer.

Figure 2. Surface morphology of aluminum nitride films.
We can distinguish between grain structure with a grain size of 200–400 nm, between the grains can be distinguished boundary dislocations, which could be formed during the growth of the embryo at the stage of early islet growth of the film, which then forms a columnar structure, sprout to the surface parallel to the axis of growth.

**Conclusion**

Magnetron sputtering makes it possible to obtain A1N films with an ordered structure, similar in structure and properties to single-crystal films, under nonequilibrium conditions.

X-ray diffraction analysis showed that the aluminum nitride films grown by this method have the orientation of the structure (002) and (111).

Depending on the technological conditions of film formation, the degree of their crystallinity varied from 10 to 30%, with a lower argon flow and a decrease in pressure, the degree of crystallinity decreases.

The influence of the parameters of the deposition process (the pressure of the gas mixture, the power of the direct current magnetron discharge scattered on the aluminum target, the concentration of argon and nitrogen) on the parameters characterizing the crystal structure of the AIN films is considered. Presents data on the preferential orientation of the axis of the texture relative to the surface normal of the substrate, the lattice parameters, size of coherent scattering regions of the orientation of the axes relative to the lattice microstrain.

Correlation analysis between the values of the power dissipated in the target, the working pressure and the size of the crystallites, and the degree of crystallinity of the films of A1N, did not reveal any relationship between these parameters, which allows us to conclude that the texture of the films is influenced by the morphology of the substrate surface, which is responsible for the size of the crystallites and the predominant orientation of the texture axis, as well as the microdeformation of the lattice, which leads to a decrease in the size of the crystallites.

The value of the relative micro deformation of the lattice ($\Delta d/d$) depending on the parameters, ranged from $1.7 \times 10^{-3}$ to $6.9 \times 10^{-3}$, with an increase in the micro deformation, the size of the CSR decreased from 29 to 8 nm.

**References**

[1] Jones D, French R, Mullejans H et al. 1999 *J. Mater. Res.* **14**(11) 4337–4344.
[2] Balasubramanian S, Belucci S, Cinque G et al. 2006 *J. Phys.: Cond. Matt.* **18** S2095-S2104.
[3] Loughin S, French R, Ching W et al. 1993 *Appl. Phys. Lett.* **63**(9) 1182–1184.
[4] Herzog T, Walter S, Hillmann S et al. 2012 Aluminum nitride thin films for high frequency smart ultrasonic sensor systems *Proc. 18th World Conf. on non-destructive testing* (16-20 April, Durban, South Africa) 21–28.
[5] Ager-III J, Wu J, Yu K et al. 2004 Group III-Nitride alloys as photovoltaic materials *SPIE proceedings of the 4th International Conference on Solid-State Lighting* 308–315.
[6] Sushentsov N, Ermolaev A, Markov A et al. 2014 Thin films in sensors on surfactants *Mat. Kharkiv Scientific Assembly ISTFE-14* 256–258.
[7] Sergeeva O, Solnyshkin A, Kiselev D et al. 2019 *Solid State Physics* **61**(12) 2386 – 2391.
[8] Kaltiev H, Sidelnikova N, Nizhankovsky S, Danko A et al. 2009 *Phys. and Tech. of Semiconductors* **43**(12) 1650–1654.
[9] Redkin A, Ryzhova M, Yakimov E, Roshchupkin D 2016 Properties of the textured polycrystalline films of aluminum nitride and zinc oxide obtained by the method of chemical vapor deposition *Proc. of the Int. Scientific and Tech. Conf., (November 21-25, Moscow) part 2* 73-77
[10] Butashin A, Muslimov A, Kolymagin A, Levachev A et al. 2017*App. Phys.* **5** 87–91.
[11] Gorelik S, Skakov Yu, Rastorguev L 2002 Radiographic and electron-optical analysis Textbook for universities (Moscow, MISIS) 360.

[12] Selivanov V, Smyslov E 1994 Factory laboratory 60(2) 31–36.

[13] Shtolz A, Medvedev A, Kurbatov L X-ray analysis of micro stresses and the size of coherent scattering regions in polycrystalline materials Educational electronic text edition. Publishing house of GOU-VPO UGTU-UPI, p 23.

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