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AlN thin film deposition using a radio-frequency beam assisted pulsed laser deposition

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Aluminium nitride is one of the compounds intensively studied because of its attractive properties: large direct bang-gap, large thermal conductivity, high acoustic velocity, high melting point, high Knoop hardness, low dielectric loss. Pulsed laser deposition was found to be a very attractive technique for obtaining films with desired properties for different applications: AlN layers can be obtained by ablation of a pure Al target in reactive atmosphere containing nitrogen and argon. The influence of the deposition parameters (gas pressure, laser fluence, Radio Frequency power) on the structure and morphology of the deposited layers was studied. Laser plasma and RF beam were characterised by optical emission spectroscopy: a correlation between the species concentration and film composition was done.

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1. Introduction

Aluminum nitride (AlN) material has many attractive properties, such as chemical and thermal stability, high thermal conductivity, high dielectric permittivity, piezoelectricity with high acoustic wave velocity and mechanical hardness. Thin films of AlN with definite microstructure are essential as electrical insulating and passivating layers and for many applications in surface acoustic wave (SAW) and UV optical devices. Several deposition techniques have been developed including rf magnetron sputtering [2,3], metal organic chemical vapour deposition (MOCVD) [4], molecular beam epitaxy (MBE) [5], filtered cathodic vacuum arc (FCVA) [6] and pulsed laser deposition (PLD) [7,8].

Throughout out of this work we report on the optical emission measurements of the laser plume and RF plasma beam and on the compositional and structural properties of the AlN thin film.

2. Experimental set-up

Radio frequency beam assisted Pulsed Laser Ablation was used as a deposition technique. The addition of a RF (13.56 MHz) plasma beam working in nitrogen/argon was founded to improve the quality of film and to reduce the amount of oxygen in film. The experimental set-up is presented in figure 1. A SURELITE II Nd:YAG laser working at a repetition rate of 10 Hz and having a width pulse of τ = 5 ns, with the possibility of changing the wavelength by frequency doubling up to the fourth harmonic (therefore λ =1064, 532, 355, 266 nm) will be used for the targets ablation. The pure Aluminium target was mounted on the rotation-translation target holder for avoiding fast drilling and having in every moment a “fresh surface” during ablation process. In all the experiments the distance target-substrate is 4cm.

Optical Emission Spectroscopy analyses were obtained using a SM-240 CCD Optical Multichannel Analyser. The light of the plasma plume and RF beam was focused by a quatz convergent lens on the entrance slit of spectrograph. The spectrograph SM-240 is based on crossed Czerny-Turner configuration, the CCD was syncronized with the laser pulse and the integration time was 10 ms.

The pressure (in the ratio of 1:2) varied in the range of 0.05 to 0.1 mbar and the laser fluence between 2 and 5 J/cm^2. A particular case was also investigated, when a radiofrequency beam was generated in the gas mixture for increasing the nitrogen reactivity. The substrates were heated during thin films deposition up to 600^{0}C – 700^{0}C.

In the present work, AlN thin films have been deposited by Radio-Frequency beam assisted Pulsed Laser Deposition. Substrates as Pt, MgO and Si were used. The nitrogen/ argon pressure (in the ratio of 1:2) was varied in the range of 0.05 to 0.1 mbar.

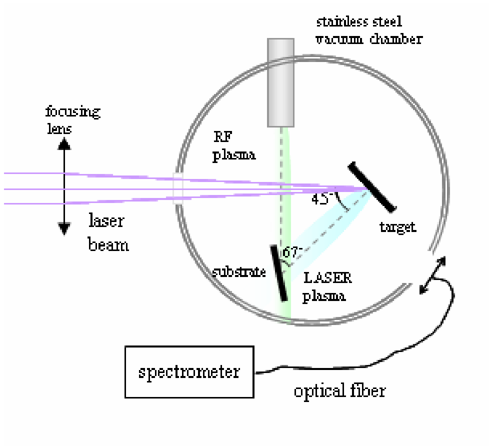

Fig. 1. The scheme of experimental set-up consist of laser, reactor chamber, RF plasma beam, target and substrate.

3. Results

The emission spectra were recorded within the range of 250-800nm. The spectra contain the plasma plume and the RF beam ignited in nitrogen and argon. The main species appear in the spectra are ions and atoms of Aluminium and strong molecular bands of nitrogen. In figure 2 is presented the emission spectrum of laser plasma. One may see that close to the target the main species of the plasma plume are atoms and ions of Aluminium.

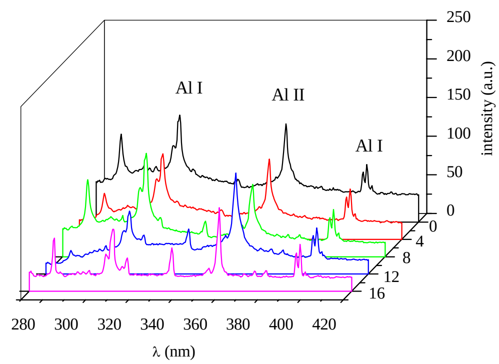

Fig. 2. Emission spectra of the plasma plume measured at different distances moving away from the Al target surface to the edge of the plasma along a normal direction.

In the emission spectra of the laser plume and RF beam lines related to oxygen were not detected. Figure 2 shows the spectra recorded by moving the entrance slit of the optical fiber from the AlN target surface the edge of the plasma plume along a normal direction with a step of 4 mm. The black spectrum was recorded at 0 mm away from the Al target. As is seen, the intensity of the peaks of Aluminium significantly depends on the distance from the surface of the target to the edge of the plume.

The main emitting species in the laser produced plasma are excited neutral Al (Al I) atoms or single Al^+ (Al II). The lines with the highest intensity in the plasma spectrum are for Al (Al I at 308.42, 309.61, 396.15 and 394.4 nm) and Al^+ (Al II at 358.6 nm).

Continuum emission spectra, shown in figure 2, centered at wavelength around 310 nm were observed close to the target. These continuum spectra shifted to the longer wavelengths (centered at 500 nm) suggesting the decrease of the plume temperature along a direction normal to the target since the shorter the light wavelength, the higher the temperature and, therefore, the higher of the photon energy is.

The emision of the N_2 second pozitive system ($C^3\Pi_u \rightarrow B^3\Pi_g$), with band heads at (0-1) 315,9 nm, (0-0) 337 nm, (1-0) 357,6nm, (2-0) 380,4 nm and the first system positiv of N_2 at 618.9nm (0-4) and 563.2 nm (0-5) are shown in figure 3.

The upper spectrum is measured at 5 mm away from the substrate, presented the main species of molecular nitrogen.

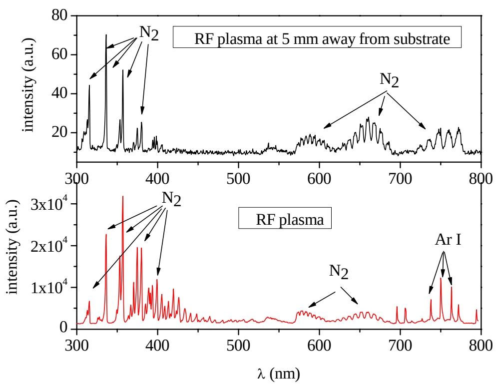

Fig. 3. Emission spectra of the plasma plume and RF beam discharge recorded at 25 mm away from the Al target (the black spectrum) and that recorded at 5mm to the substrate (the red spectrum).

In Fig. 3 the lower emission spectra of the nitrogen is presented between target and substrate at 20 mm. In this spectrum, positions of Ar I atomic lines at 738.39 nm, 750.39 nm and 763.51 nm are presented.

Molecular AlN was not observe in the emission spectrum, therefore we suppose its formation on the surface of the substrate. The presence of excited atoms and ions of Al are not presented near to the substrate maybe being under limit of detection of our spectrograph. Therefore we suppose that most of the species existent in the plasma as (Al I, Al II, N_2, N) stick at the surface of substrate.

It was observed that increasing the laser fluence from 2 to 5 J/cm^2 the intensity of Al lines increases and aluminium metallic could be detected on the deposited layer, coming directly from the target. The optimisation of the fluence getting the AlN in the thin film is arround 2 – 3 J/cm^2, under our experimental conditions.

The crystalline status and the composition of the deposited layer obtained in the experiement were studied by the XRD diffraction. These analyses were conducted with a Szimadzu 6000 difractometer. The investigations were carried out with θ-2θ configuration.

Typical XRD diffraction pattern of the deposited layer is presented in figure 4 for an AlN layer on Pt substrate. The nitrogen/argon pressure is 0.5 mbar. The peak at 33^o is assign to AlN <100> hexagonal phase, with the rest of the peaks coming from the substrate.

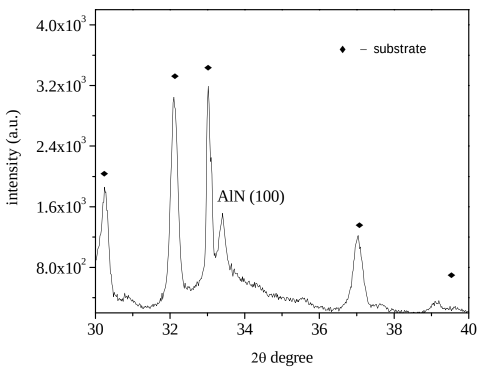

Fig. 4. XRD patterns of AlN films deposited at N_2/Ar atmosphere, gas pressure of 0.5 mbar on a Pt substrate. The substrate temperature was 700^{o}C and the RF power 150W.

A Nomand microscope, from Quesant, in the intermittent contact mode for observing the surfaces of the thin films has been used. The Atomic Force Microscopy investigations showed compact surfaces, without droplets and cracks and small surface roughness (fig. 5). The RMS value on the AFM surface measurements is 18 nm.

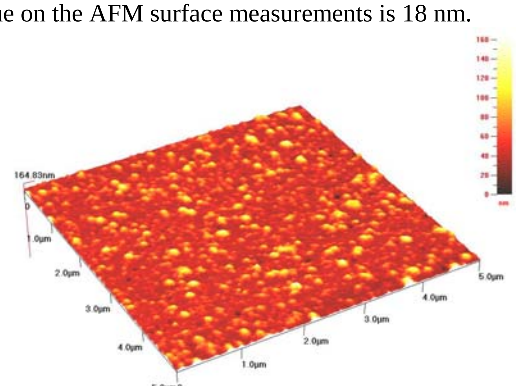

Fig. 5. AFM image of the AlN film deposited on Pt, in nitrogen and argon atmosphere pressure 0.1mbar, substrate temperature 700^{0}C and RF power 150W.

The roughness of the AlN deposited on different substrate is around 20-50 nm and slightly dependent on the experimental conditions. At high power of RF plasma the roughness increases especially due to possible droplets formation on the substrate. From the ruoghness point of view the optim of the RF power is 50-70 Watt.

4. Conclusions

Optical emission measurements were used to get the information of the plume and RF beam plasma composition close to the target and near to the substrate. No detection of molecular AlN was observed in our plasma. It was found that increasing the laser fluence metallic Al could be finding in the deposited layer. AFM showed a higher roughness, compact and without droplets surfaces.

Acknowledgements

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