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Full Length Article

Deposition of nanostructured crystalline alumina thin film by twin targets reactive high power impulse magnetron sputtering

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ABSTRACT

In order to suppress the arcs in reactive sputtering deposition process, twin targets reactive high power impulse magnetron sputtering (TTR-HiPIMS) was proposed, and alumina (Al_2O_3) thin films were synthesized on silicon (1 0 0) substrates by this method under various argon/oxygen gas flow rates and substrate temperatures. The deposition rate of alumina films decreased from 110 to 85 nm/h when the oxygen flow rate was increased from 8 to 16 sccm. The elemental composition of the films was analyzed by both energy dispersive spectroscopy and X-ray photoelectron spectroscopy, and the results revealed that the suitable argon/oxygen gas flow rate for depositing a stoichiometric alumina film was 50/14. Grazing incidence X-ray diffraction (GIXRD) results indicated that all the as-deposited films were polycrystalline γ-Al_2O_3. The GIXRD patterns also confirmed that under optimum process conditions, crystalline alumina films can be obtained at temperatures as low as 300 °C, and the intensity of the diffraction peaks increased with the substrate temperature. Scanning electron microscopy showed that all the films have a smooth surface, which indicated that the arc events were almost completely suppressed in the sputtering process. The films prepared at substrate temperatures ranging from 300 to 500 °C contained fine nanocrystals of sizes in the range 15–25 nm. Furthermore, atomic force microscopy results revealed that the root-mean-square roughness of the films increased from 2.06 to 4.24 nm when the substrate temperature was raised from 300 to 500 °C. The results suggest that the new developed TTR-HiPIMS technique is a simple and effective method for the reactive deposition of nanostructured crystalline alumina films at relatively low substrate temperature.

1. Introduction

Owing to their excellent properties, such as wide bandgap, high transparency, large hardness, high wear resistance, excellent dielectric properties as well as good chemical and thermal stability, alumina (Al_2O_3) thin films have received increasing interest in different industrial applications from optoelectronic devices, medical insets and wear resistant coatings to catalysis applications [1–4]. All these applications, especially for optical anti-reflection coating, require the deposited alumina films have good homogeneity, low surface roughness and proximity to stoichiometry [5]. Furthermore, compared with its amorphous form, many of the desirable properties are tied in with the crystalline phases of alumina [6]. Alumina has 24 known crystalline phases, among which the α-Al_2O_3 is the only thermodynamically stable phase, other important and commonly observed metastable phases are γ-, κ-, θ-, and δ-phases [7]. The existence of this polymorphism, however, complicates the growth process of a particular phase or phase mixtures.

Various technologies, such as magnetron sputtering [8,9], filtered cathodic vacuum arc technology [10], electron beam evaporation [3,11], plasma spraying [12], plasma-enhanced chemical vapor deposition [13] and sol-gel process [14], have been used to deposit alumina thin films. However, growth of crystalline alumina thin films by these methods at low temperature is usually difficult. Previous studies have shown that the crystalline alumina could be fabricated at relatively low temperature by combine the normal sputtering equipment with other ion sources, for example, rf coil and magnetic field arrangement near the substrate holder, to enhance the mobility of near-surface species via low energy ion bombardment [15,16]. But these methods increase the cost and complication of the equipment. Deposition on Cr_2O_3 templated layer can also result in the growth of crystalline alumina films at reduced temperature [17]. But the pre-deposited layer increases the structural complication of the deposited film system, which is not desired.

High power impulse magnetron sputtering (HiPIMS), which was introduced by Kouznetsov et al. [18] in 1999, is a novel and promising

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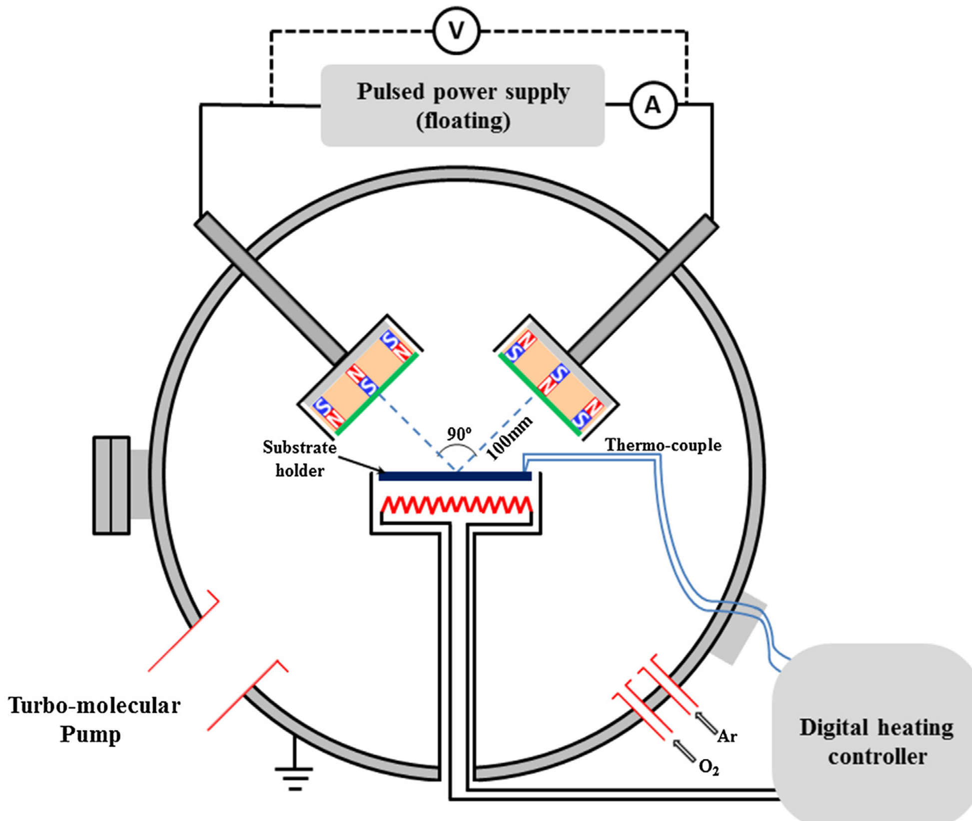

Fig. 1. Schematic diagram of the experimental set-up.

ionized physical vapor deposition technique that allows for growth of films with superior properties. In HiPIMS, power is supplied to the target (cathode) in unipolar pulses of high magnitude but low duty cycle (< 10%) and frequency (< 10 kHz) leading to pulse target power densities on the order of kW/cm^2, while maintaining the time-averaged target power in values similar to those during dc magnetron sputtering [19,20]. This mode of operation results in generation of ultra-dense plasmas with unique properties, such as a high ionization fraction of sputtered atoms and high ion fluxes to the substrate [21]. In addition, HiPIMS also presents the advantage of either reduce the hysteresis effects in reactive sputtering, or at least improve the process stability in the transition mode [22,23]. However, sputtering pure metallic aluminum target in a reactive oxygen environment under conventional HiPIMS conditions, for example, single target discharge, pulse length in the order of tens of microseconds and repetition frequency up to 4 kHz, always lead to the build-up of a thin, electrically insulating alumina thin film on the uneroded areas of target surface, which could cause severe arcing and target or power unit damage [24]. Moreover, deposition of dielectric alumina thin film on the substrate is always accompanied by the coverage of all inner surfaces of the vacuum chamber, which results in drifting potentials, unstable deposition process, and consequently, undefined film properties [25]. In addition, reactive RF magnetron sputtering allows realizing alumina thin films with arc events being greatly suppressed, but the complex power circuit and scaling problems limit its manufactory industrial uses [26,27].

In this study, alumina thin films were prepared by a new developed twin targets reactive high power impulse magnetron sputtering (TTR-HiPIMS) technique under different deposition conditions. With this system, the periodic target voltage reversals make it possible to remove the accumulated charges from both targets, and hence suppress arcing and stabilize the reactive sputtering process [28]. Additionally, the high power pulsed plasma combined with the twin targets configuration can lead to a significant ion bombardment of the growing film and consequently an enhancement of the film properties.

2. Experimental

Alumina films were synthesized in a twin targets sputtering system designed by ourselves, as shown in Fig. 1. The well-cleaned single crystal silicon (1 0 0) wafers ($25 \times 25 \times 0.7\ mm^3$) with a native oxide layer on top was used as substrate. The system was equipped with two circular aluminum targets (99.999% purity) of 100 mm diameter and 6 mm thickness, each tilted at an angel of 45° from the substrate holder axis and the target-to-substrate distance was 100 mm. Prior to the deposition, the vacuum chamber was evacuated with a turbo-molecular pump to a base vacuum of better than 1×10^{-3} Pa. The single crystal silicon (1 0 0) substrates was thoroughly cleaned consecutively in an ultrasonic bath with acetone, ethanol, then rinsed with deionized water and dried by an air blow device. Both targets were pre-sputtered in pure argon atmosphere for 20 min to remove contaminants, if any on the targets surface. For the deposition process, both argon and oxygen were fed into the chamber through injectors and their flow rates were monitored individually using mass flow meters. The argon flow rate

Table 1
Main processing conditions for the deposition of different thin film samples.

Sample no.	Argon flow rate (sccm)	Oxygen flow rate (sccm)	Substrate temperature (°C)	Deposition time (min)
S1	50	8	500	60
S2	50	11	500	60
S3	50	14	500	60
S4	50	16	500	60
S5	50	14	300	60
S6	50	14	400	60
S7	50	14	450	60