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Multifractal spectra of atomic force microscope images of lanthanum oxide thin films deposited by electron beam evaporation

Guodong Liu [a,b,*], Lingyuan Wu [a,b], Fuping Zhang [a]

[a] *Institute of Fluid Physics, CAEP, Mianyang 621900, China*
[b] *Key Laboratory of Science and Technology on High Energy Laser, CAEP, Mianyang 621900, China*

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ABSTRACT

The concept of fractal geometry has been widely used in describing structures and processes in experimental systems. In this letter, La_2O_3 thin films were deposited by the electron beam evaporation on a silicon substrate and then were annealed at 900 °C for various times. The surface characteristics of the films were investigated by means of an atomic force microscope method. The relationship between the time of the rapid thermal annealing process and the characteristics of the resulting surface morphology was examined. Multifractal spectra $f(a)$ show that the effect of annealing time on the width of spectrum and Δf is apparent. The various multifractal spectra indicate that the surface roughness decreases firstly and then increases with increase in the annealing time. These results show that the atomic force microscope images can be characterized by the multifractal spectra.

1. Introduction

Rate earth oxides with high relative dielectric constant ($k \geq 20$) such as Ta_2O_5, HfO_2, ZrO_2, Pr_2O_3, Yb_2O_3 and La_2O_3 are currently being investigated as potentials replacements for SiO_2 in the next generations of complementary metal–oxide–semiconductor (CMOS) devices [1,2]. The underlying idea is that high-k insulators allow physically thicker films to achieve a combination of required capacitance value and acceptable leakage current. This has two major consequences for transistors: firstly, improvement of the channel current control and secondly, diminishing of the standby power dissipation, an issue of utmost importance for low-power applications. Among the referred material [3–6], lanthanum oxide (La_2O_3) is expected to give better performances owing to a large energy band gap (5.5 eV) [7], high electrical breakdown field strength [8], high dielectric constant (k=27) [9] and better thermodynamics stability on Si substrate [10]. Moreover, La_2O_3 has been shown to be compatible with current semiconductor manufacturing processes [11,12], a critical factor both from the technological and economical viewpoints.

Some studies have been performed on electrical and microstructural characteristics of gate dielectric of La_2O_3 ultrathin films [13–18]. Furthermore, it has been reported that surface morphology plays an important role in the thin film electron transport [19,20]. However, a few investigations have been made about the influence of post-processing on the surface morphologies of La_2O_3 thin films. In this letter, the La_2O_3 thin films were deposited by electron beam evaporation (EBV) and then the as-deposited films were annealed using the rapid thermal annealing (RTA) process to

* Corresponding author at: Institute of Fluid Physics, CAEP, Mianyang 621900, China.
E-mail address: gd.liu123@sohu.com (G. Liu).

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improve the surface roughness. The surface topographies of the La_2O_3 thin films annealed at 900 °C for different times were measured by an atomic force microscope (AFM). Usually, surface roughness conditions are characterized by atomic force microscopy (AFM) with use of rms roughness values. Rms roughness describes the long-range average amount of rise and fall relative to average height. It cannot express directly the maximum range of particle height distribution. Besides, rms roughness values often change with resolution.

It has been recently realized that concepts from fractal geometry may be used to describe the surface morphology and complexity of thin films. The fractal geometry is a branch of modern mathematics that uses fractional dimensions to describe disordered objects in which some possess non-integer dimensions. When a surface topography or nanostructure will reach a non-integer dimension, a fractal analysis might be used to study the surface morphology of nanostructure [21]. In this situation, fractal dimensions greatly simplify the descriptive properties of surface morphology. In contrast to traditional analyses, the fractal analyses can be used to extract different types of information from measured surfaces. This makes fractal analysis widely applicable and very useful in describing complex surface characteristics of thin films and in advancing our understanding of how the geometry of surfaces affects the physical properties of the system. Moreover, fractal analysis can express directly the maximum range of surface height distribution and is independent of the change in resolution. In the past few years, especially the multifractal spectra $f(a)$ in fractal analysis have been found to have very important application in electron microscopy. Multifractal spectra are used not only to characterize the surface roughness but it can also be used to characterize the shape of peaks and valleys between different rough surfaces. Yadav et al. applied it to quantitatively investigate the structural properties of LiF thin film surfaces [22]. Yu et al. performed multifractal spectra to analyze the surface topographies of amorphous electroless Ni–Cu–P alloys [23]. Sun et al. used multifractal spectra to characterize the local growth probabilities of ZnO films deposited by a reactive sputtering method [24]. Apparently, the multifractal provides a framework that can quantify the structural complexity of a vast range of physical phenomena. Therefore, we present multifractal spectra of La_2O_3 thin films to characterize the surface roughness for various annealing times in this study. The results indicated that the RTA conditions greatly affect the final surface roughness of La_2O_3 thin films.

2. Experimental and multifractal calculation

The La_2O_3 thin film of 12 nm thickness was prepared by the EBV method. La_2O_3 granule (purity 99.99%), between 3 and 5 mm in diameter, was used as the starting material. The evaporated substance was collected on a Si(100) substrate, which was mounted on a substrate holder 50 cm away from the evaporation source. The substrate temperature was set at 200 °C. The thickness and the deposition rate were controlled to be as 12 nm and 0.5 Å/s using a quartz crystal monitor. The high vacuum in the deposition chamber was achieved using a vacuum diffuse pump. The base pressure prior to evaporation was about 2×10^{-3} Pa, and the working pressure during evaporation was about 8×10^{-3} Pa. After the deposition of the La_2O_3 thin films, the RTA process was carried out for 1, 5 and 10 min at 900 °C in order to decrease the surface energy of the films via thermodynamic mechanisms and minimize irregularities of the film surface. Furthermore, the RTA process tends to restrict the film crystallinity. Finally, an AFM apparatus (SPM-9500J2, Shimadzu, Tokyo, Japan) was used to scan the specimens in order to measure the morphological properties of the La_2O_3 thin film surface. Multifractal analysis of the AFM image was carried out for $0.5\times0.5\ \mu m^2$ scan areas.

The AFM images are gray-scale digital pictures of 512×512 pixels. Each gray-scale value corresponds to a height of the film. Multifractal spectra of AFM images are calculated by a box-counting method. The images can be divided into many boxes of size $l\times l$, and let $\varepsilon=l/L$ ($L=512$) ($\varepsilon<1$). $P_{ij}(\varepsilon)$ is the height distribution probability of the film in the box (i, j). It is determined as

$$P_{ij}(\varepsilon)=h_{ij}(\varepsilon)/\sum h_{ij}(\varepsilon)=h_{ij}(\varepsilon)/h \quad (1)$$

where h_{ij} is the average height of the box (i, j) of size ε measured from the substrate surface, which is define as the datum planes in the calculations. $P_{ij}(\varepsilon)$ can be described as multifractal as

$$P_{ij}(\varepsilon)\sim\varepsilon^{a} \quad (2)$$

where α is the singularity of the subset of probabilities. The larger the α, the smaller the probabilities. The number of boxes of size ε with the same probability can be described as

$$N_a(\varepsilon)\sim\varepsilon^{-f(a)} \quad (3)$$

where $f(a)$ is the fractal dimension of a subset. A fractal dimension is an index for characterizing fractal patterns or sets by quantifying their complexity as a ratio of the change in detail to the change in scale. It is used as a measure of the space-filling capacity of a material pattern. The dependence of $f(a)$ on a is the multifractal spectrum. In the situation of random multifractal, the partition function method is used to calculate $f(a)$. The partition function $\chi_q(\varepsilon)$ is defined and expressed as a power law of ε with an exponent $\tau(q)$, where q is the moment order $(-\infty<q<+\infty)$

$$\chi_q(\varepsilon)=\sum P_{ij}(\varepsilon)^q=\varepsilon^{\tau(q)} \quad (4)$$

$\tau(q)$ can be obtained from the slope of $\ln\chi_q(\varepsilon)\sim\ln\varepsilon$ curve

$$\tau(q)=\lim_{\varepsilon\to0}[\ln\chi_q(\varepsilon)/\ln\varepsilon] \quad (5)$$

$f(a)$ can be obtained by performing Legendre transformation as follows:

$$a=d[\tau(q)]/dq \quad (6)$$

$$f(a)=aq-\tau(q) \quad (7)$$

The q value cannot be infinite in real calculation. The influence of $|q|$ on the multifractal spectra is more and more minor when $|q|$ increases to certain degree. $|q|$ can be considered as the maximum when $f(a)$ and a approach to the saturated values with the increase of $|q|$. Li et al. [25] took $|q|_{max}$ of 9 in the analysis of spatial distribution of the secondary electrons on a solid surface and in the bulk,