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Inherent surface roughening as a limiting factor in epitaxial cluster deposition

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Abstract

Deposition of nanoclusters at thermal energies will result in an onset of roughening of the deposited surface. In order to grow epitaxial films using cluster deposition at soft landing conditions, the effect of this inherent surface roughness on the alignment of deposited clusters must be investigated. Using molecular dynamics computer simulations we have determined the maximum size of Cu clusters that will align epitaxially, upon deposition at thermal energies, on rough (100) Cu substrates with temperatures ranging from 0 K to 750 K. We have also shown that the likelihood of epitaxial alignment for the resulting structures is dependent on the point of impact of a cluster relative to previously deposited clusters.

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1. Introduction

The use of nanocluster deposition as a means of thin film growth has been widely researched throughout the past two decades [1,2]. The leading candidate among cluster deposition techniques has long been energetic cluster impact (ECI) methods [3], due to their performance as growth processes for high quality thin films, coupled with their tendency towards facilitating a natural smoothing of the produced film. Despite this, deposition of nanoclusters at thermal energies is also a method with alluring properties.

The prospect of growing structured thin films, at conditions which are gentle towards the underlying substrate, by using pre-structured nanoclusters has facilitated the study of interactions between clusters and the surfaces upon which they are deposited. One possible application is the growth of nanocrystalline thin films, where the average grain size is of the same order in size as

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the deposited clusters. Another application is the growth of epitaxial thin films, through methods which take advantage of the crystalline structure of nanoclusters.

Thermal deposition of clusters may be one technique available for the growth of epitaxial films. It has been experimentally verified that when a small enough cluster lands on a single-crystalline substrate, it will align completely epitaxially with the substrate almost immediately after impact, i.e. achieve complete contact epitaxy [4,5]. The largest clusters that gain full contact epitaxy obey a temperature dependent upper limit in size. In a previous study, using molecular dynamics (MD) simulations we have quantitatively determined this upper size limit for Cu nanoclusters deposited with thermal energies onto smooth (1 0 0) Cu substrates at different temperatures [6].

Deposited clusters of sizes near this limit will form hillock-like structures on the substrate, thereby causing a roughening of the substrate surface. In order to grow epitaxial thin films with thicknesses exceeding one monolayer of clusters, this effect of inherent surface roughening must be accounted for in the upper size limit of clusters that will align epitaxially. In other words, one must determine the size limit for clusters that will align epitaxially when landing on a rough surface.

2. Method

Classical molecular dynamics simulations were used to simulate the deposition of Cu nanoclusters on Cu surfaces. The conditions used in the simulations did not differ much from the conditions of single cluster deposition on smooth surfaces which were described in detail in a previous paper [6]. The Cu atom interaction was described with the use of Foiles embedded-atom method (EAM) potential [7]. The Berendsen temperature control algorithm [8] with a time constant of 300 fs was used to stabilize the temperature at the borders and the bottom of the simulation cell.

The clusters were given the shape of Wulff polyhedra [9] with the dimensions of each cluster volume optimized to the configuration of minimum surface energy, as was the case for deposition of clusters on a smooth surface. In order to realistically mimic the conditions of surface roughening clusters were deposited on top of the relaxed structures resulting from previously deposited clusters. In effect, the epitaxial alignment of clusters deposited on surfaces roughened by the cluster deposition itself was examined. The orientations and positions of the deposited clusters were chosen randomly and deposition itself was done by giving the clusters a velocity in the direction of the substrate corresponding to a kinetic energy of 5 meV per atom in the cluster. The clusters were then relaxed for 2 ns before the next clusters were deposited on top of them, at positions chosen randomly with respect to the previously deposited clusters. These simulations were carried out for several different cluster sizes over a temperature range of 0–750 K.

The degree of epitaxy for the deposited cluster structures was analysed both visually and analytically. Analytical analysis was carried out by calculation of a factor we call F_{epi}, for every atom of the clusters. F_{epi} of an atom is calculated by comparing the unit vectors from each atom to its nearest neighbours, r_{nn}, to the unit vectors which are expected if the atoms were aligned with the substrate in an ideal epitaxial structure, r_{Ideal}. For every atom the dot product is calculated for all the combinations of ideal vectors and the vectors to the nearest neighbours, and the minimum value is added to the F_{epi}, i.e.

$$F_{\text{epi}} = \sum_i \min_j (\arccos |r^j_{\text{Ideal}} \cdot r^i_{\text{nn}}|).$$

The average F_{epi} of the atoms in a cluster will give an indication of the degree of strain to which the cluster is subjected. If the strain grows too high and a grain is formed, all of the atoms in the grain will contribute to a raising of F_{epi}, and hence increase the measure of non-epitaxiality.

3. Results and discussion

The interaction between thermally deposited Cu nanoclusters and rough Cu substrate surfaces does not qualitatively differ much from what was previously observed for clusters deposited on smooth