Nanostructure Design by Atomic Shadowing

by

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ABSTRACT

The objective of this research work is to develop an understanding of the atomic process, especially the roles of surface diffusion and surface pattern, that govern the formation of nanostructures by the current state-of-the-art glancing angle deposition (GLAD) technique and to explore the possibility of extending the capabilities of GLAD to Simultaneous Opposite Glancing Angle Deposition (SOGLAD) for multi-component nanostructuring.

The effects of surface diffusion during GLAD were investigated by growing periodic Ta nanopillar arrays on patterned substrates at temperatures T_s ranging form 200 to 900 ^oC. For growth at high T_s, the structure and morphology of Ta nanopillars is strongly affected by the increased surface diffusion. Branching of pillar tops occurs for growth at low T_s, while sub-pillars form during the nucleation stage at the pillar bottoms at high T_s. The formation of sub-pillars at the bottom is attributed to a transition to a multi-pillar nucleation mode at high T_s. There is also a transition to a competitive columnar growth mode at elevated temperatures, caused by an increased adatom diffusion length. The competitive growth mode results in an increase in the average pillar width, a decrease in the pillar separation and pillar number density, the accelerated growth of some pillars at the cost of others which die out, and an increased probability for the merging of neighboring pillars. The top branching is associated with kinetic roughening and limited adatom mobility at low T_s. The fraction of branched pillars decreases with increasing T_s, due to an increased lateral diffusion length, and is described with a simple nucleation model, providing an effective activation energy for Ta surface diffusion of 2.0 eV.

The surface patterning effects were studied by growing nanostructured layers onto two types of regular surface patterns: honeycomb nanodot arrays and pyramidal pit arrays, created by Nanosphere Lithography (NSL). GLAD on NSL nanodot patterns results in honeycomb nanopillar arrays while GLAD on inverted pyramid pit patterns leads to two level porous layers with microscopic pores (> 300 nm) interconnected by nanoscale (5-50 nm) pores. Statistical size analyses from honeycomb nanopillar arrays show that the distribution in *w* broadens with increasing *l* and decreasing *w*, but remains

approximately constant with a fixed *l*/*w*-ratio. This is attributed to an intercolumnar growth competition that exacerbates nanorod size fluctuations but scales with rod size, indicating that the overall nanostructure-shape during low-temperature GLAD is independent of material-specific length-scales and, therefore, completely controlled by the geometric shadowing. Microstructural evolution of two level porous Ta and Al GLAD layers is dominated by long-range atomic shadowing interactions that lead to the formation of separated vertical rods and the development of a bimodal pore structure. The nanorods grow in a competitive mode which causes rod broadening and rod extinction. Both the broadening and extinction of the rods can be quantified by a power law. The large surface diffusion length for Al rods causes a lateral growth which enhances the rod broadening and, in turn, leads to the close-up of the microscopic pores, indicating that Al layers grown on pit arrays approach a microstrostructure that is comparable to GLAD layers grown on flat substrates. In contrast, the bimodal pore structure is well preserved during Ta growth, which indicates that substrate patterning is a useful approach to create complex pore arrangements in GLAD layers for growth conditions where surface diffusion is negligible.

GLAD has also been extended to SOGLAD to fabricate novel Si/Ta two component nanostructures onto self-assembled close-packed silica nanosphere arrays. The two component nanostructures are shaped into zigzags or nanopillars by adjusting the deposition angle and/or the substrate rotation. By manipulating the sequence of the deposition, that is, by sequential or simultaneous deposition from two sources, complex nanostructures are formed where the two components are stacked vertically, laterally, or in a checker board arrangement. Scanning electron microscopy, back scattered imaging, and transmission electron microscopy provide clear compositional and microstructural contrast and show sharp (< 30 nm) vertical and horizontal interfaces. The results show that nanostructures can be designed and created by exploring the shadowing effect during glancing angle deposition. These novel nanostructures are expected to have complex functionalities and physical properties.