

DOE Final Report for grant DE-FG02-02ER46011.

Fundamental Investigations of Nanoscale Phenomena in Beam-Assisted Nucleation, Growth and Surface-Smoothing.

Principal Investigator: C Peter Flynn

Address of Institution: Board of Trustees, University of Illinois at Urbana
Champaign, Urbana, IL 61801.

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Final Report.

The purposes for which this grant was provided were specifically (1) to construct a tandem instrument that combined a low energy electron microscope (LEEM) with an ion beam source capable of irradiating a sample during observation of the surface using LEEM; and (2) to employ the new machine to whatever degree possible to observe the evolution of clean crystal surfaces during ion beam irradiation. A principal motivation was to investigate the fundamental behavior of radiation damage under circumstances for which the damage can be observed directly in real time as it occurs. A second main motivation was to create tunable perturbations of the defect (adatom and advacancy) equilibrium on clean crystal planes and in this way explore the fundamental kinetics of surface behavior that enters into numerous phenomena of interest to DOE including surface erosion, catalysis, and the damage to crystals caused by impacts of energetic particles.

The funding has been employed to successfully pursue all the original goals, and additional opportunities that developed as a result of discoveries made in this research. A summary follows.

1. Construction. In initial analysis it became clear that the LEEM is compatible only with a source of negative ions because the sample to be irradiated is at -15 keV. Accelerators of this type are rare but a suitable source, the SNICS II, was obtained from National Electrostatics Co of Madison Wis. and modified for improved vacuum and focus. The source was integrated into the vacuum of the LEEM without loss of vacuum integrity, and developed to give fluxes up to 0.1 ML/s of the desired ion species on the sample. In order to simplify the surface processes to interpretable form, use typically was limited to self-ions (eg, Pt⁺ on Pt(111)).

2. Research Accomplishments. The new instrument has been employed in a variety of fundamental research initiatives made available by the new capabilities. Major new results include, but are not limited to, the following:

A. Synthesis. A beam of self-ions has been used to synthesize a number of novel surface structures that have valuable research applications. These include the

synthesis of pans and mesas comprising perfect terraces $\sim 5\mu\text{m}$ in diameter for use as arenas for designed experiments (see below). Fourier waves have been created on step edges for measurements of diffusion rates (see below). Islands of both types (adatoms and advacancies) have been created for fundamental investigations of nucleation and of growth.

B. Fundamental Investigations. The decay with time of Fourier waves synthesized on step edges have been used to determine the surface mass diffusion coefficient over an unprecedented range of $\sim 10^5$. Surface 'clocks' in the form of growth spirals and Bardeen-Herring sources have been employed to determine the absolute efficiencies with which energetic collisions create surface defects, for comparison with molecular dynamics calculations. We have used a beam of self-ions to drive pans and mesas off equilibrium until a new island is nucleated, and have documented the conditions under which irradiation causes nucleation for comparison with fundamental theory. In examining the subsequent driven growth of ions we have discovered a universal behavior previously not recognized and have explained the observations using a new theory (see below).

C. Theory. As these investigations developed it became apparent that existing theory of surface kinetics was not adequate to describe the phenomena under study. A comprehensive treatment of surface kinetics including reactions among surface thermal point defects (adatoms and advacancies) was undertaken successfully. The results are a critical factor in the quantitative interpretation of the experiments on diffusion, nucleation, growth and driven clocks described in B above.

Summary and Future Prospects. The LEEM-SNICS II tandem has been built and successfully applied to important initial problems. The research using this new tool is just begun, with many opportunities still not addressed. The opportunities for new efforts employing the time domain for detailed kinetic studies appear particularly promising.

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