HARNESSING PETASCALE COMPUTING TO EXPLAIN FUNDAMENTAL MECHANISMS DRIVING NANOPATTERNING OF MULTICOMPONENT SURFACES BY DIRECTED IRRADIATION SYNTHESIS

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EXECUTIVE SUMMARY

Ion beam-driven synthesis of quantum dots on III–V semiconductor surfaces is a promising approach toward a device-scale fabrication process for solar cells, quantum dot lasers, and other applications. However, the physical mechanisms that determine the pattern formation and properties such as quantum dot spacing remain poorly understood. To close this knowledge gap, massive-scale molecular dynamics simulations have been performed studying ion beam bombardment of gallium antimonide (GaSb) surfaces with 500 eV Ne[+], Ar[+], and Kr[+] ions. Incident energetic ions are able to induce the formation of single-element clusters within the GaSb surface, with Sb clusters being far more prevalent than Ga clusters. These clusters may provide the basis for long-timescale mechanisms beyond the scope of molecular dynamics, such as radiation-enhanced diffusion, to create the compositional depth profile observed in experimental studies.

INTRODUCTION

The ability to synthesize unique nanostructures and nanopatterns at surfaces by ion beam bombardment holds great potential as a scalable technique to create novel nanomaterials in a single process step. In particular, the ability to fabricate quantum dots on III–V semiconductor surfaces, such as Galium Asenide (GaAs) or Gallium Antimonide (GaSb), using ion beam irradiation could be used to efficiently functionalize those materials for use in high-efficiency solar cells or quantum dot lasers. However, since the first discovery of these nanodots [1], theoretical models have been unable to provide a complete, predictive description of the nanopattern formation on these surfaces. Consequently, the dependence of nanopattern formation on III–V semiconductor surfaces on experimental parameters such as the incident ion mass or energy remains poorly understood.
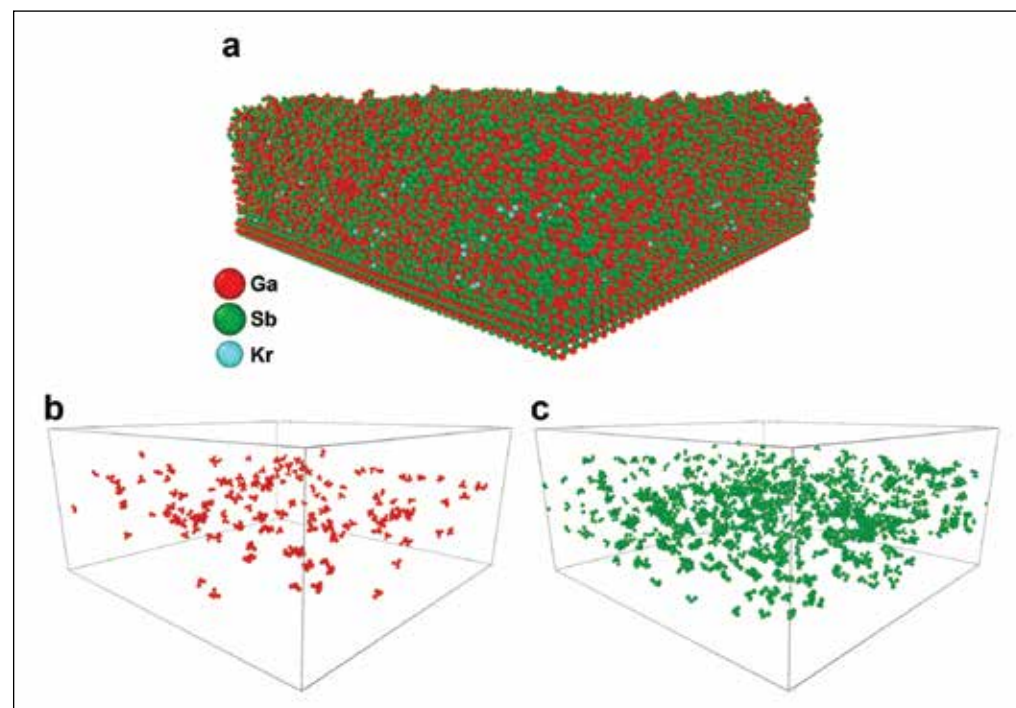

Figure 1: Snapshots of the GaSb surface after irradiation by 500 eV Kr[+] ions to a fluence of 7.5×10^{15} cm^{-2}—(a) snapshot of the entire surface after ion bombardment, showing the lack of any notable compositional depth profile; (b) snapshot showing only Ga atoms in clusters; (c) snapshot showing only Sb atoms in clusters. Sb atoms show a significantly greater tendency to form clusters within the irradiated GaSb surface.

To address this knowledge gap, atom-level computational modeling is necessary to observe and characterize the ion-induced changes in the surface as they occur, providing detailed insight into the ion-surface interactions at length and time scales far smaller than what can be experimentally observed. At the same time, any simulation efforts must also be able to connect the small-scale atomistic mechanisms to the evolution of the larger surface at experimentally observable length and time scales. Determining both the large and small scales of ion-driven nanopatterning can only be done with high-performance computational power on a massive scale, well beyond that provided by conventional computing platforms.

In previous work conducted on Blue Waters [2], massive-scale molecular dynamics simulations were conducted to connect experimental observations of the compositional depth profile [3,4] to pattern-forming surface instabilities. The results showed, for the first time, that an ion-induced compositional depth profile led to lateral phase separation, providing a pathway for the pattern-forming instability to emerge. The present simulations seek to elucidate the ion-induced mechanisms causing the formation of the compositional depth profile that drives these changes.

METHODS & RESULTS

Molecular dynamics (MD) simulations of ion bombardment are carried out with the LAMMPS package [5] for 500 eV Ne[+], Ar[+], and Kr[+] ion species incident on an initially pristine GaSb surface. The simulation cells were 25 × 25 nm2, providing sufficient space for lateral compositional variations to evolve on the same scale observed in previous simulations. For each ion species, the GaSb surface was irradiated to a fluence of 7.5×10^{15} ions/cm^2, an experimentally relevant fluence (the sum of the energies of the particles per unit area contained in the particles with which a material is irradiated) on the same order of magnitude as the threshold fluences for pattern formation that have been observed in experiments.

Snapshots of the resulting surfaces after ion irradiation are shown in Fig. 1 for the 500 eV Kr[+] case. Three major observations can be made from these snapshots. First, despite irradiation to a fairly large fluence, no compositional depth profile was observed. This indicates that additional long-temporal scale mechanisms are necessary to accurately model the compositional evolution of the surface, such as radiation-enhanced diffusion or stress-driven flow. Second, while a depth profile does not emerge, the formation of smaller clusters of Ga or Sb can still be observed due to the prompt ion effects. This indicates that even at short timescales the energetic ions still induce compositional changes in the surface, which may provide "seed" structures to grow into larger-scale lateral compositional gradients. Finally, Sb atoms show a much stronger clustering tendency than Ga atoms, which may be indicative of the higher mobility of Ga compared to Sb seen in experimental studies [6].

Interestingly, different ion species were shown to have differing effects on the surface compositional evolution, as shown in Fig.
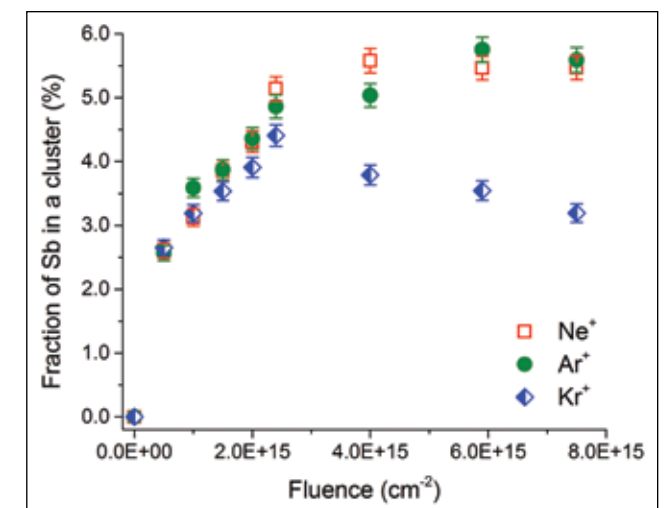

Figure 2: Fraction of Sb atoms which are members of clusters as a function of increasing fluence for each ion species. While all three ion species initially induce similar clustering behavior, for fluences greater than 2.4×10^{15} cm^{-2} Kr[+] irradiation causes a decrease in clustering compared to the lighter two ion species.

2. In the early stage of ion bombardment, the tendency for Sb clusters to form is similar for all ion species. However, at higher fluences the Sb clustering tendency decreases for Kr[+] irradiation, while it increases asymptotically for the lighter Ne[+] and Ar[+] ions. While the implications of this observation are still unclear, the difference correlates well with the experimentally determined threshold fluences for nanopattern formation, which is a factor of four greater for Ne[+] and Ar[+] ions compared to that for Kr[+] ions [4].

WHY BLUE WATERS

This work required carrying out three MD simulations in parallel, each of which contained around 150,000 to 200,000 atoms at any time, for over 230 million time steps each. On a conventional computing cluster, this would be practically impossible and would require several years to complete, at best. Furthermore, several terabytes of data were generated from these simulations, necessitating a high-performance parallel file I/O system to minimize the impact on the overall computation time.

PUBLICATIONS AND DATA SETS

Lively, M. A., B. Holybee, M. Toriyama, and J. P. Allain, Massive-scale molecular dynamics of ion-irradiated III–V compound semiconductors at the onset of nanopatterning, *Nucl. Instrum. Meth. Phys. Res. B* (2017), in press.