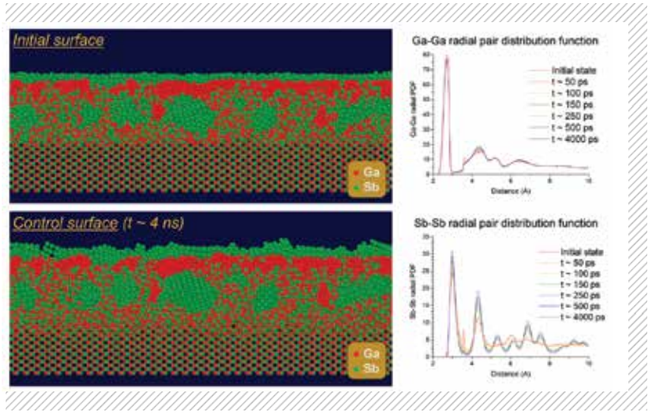
HARNESSING PETASCALE COMPUTING TO ELUCIDATE FUNDAMENTAL MECHANISMS DRIVING NANOPATTERNING OF MULTICOMPONENT SURFACES BY DIRECTED IRRADIATION SYNTHESIS

Allocation: Illinois/50.0 Knh
PI: Jean Paul Allain[1]
Collaborators: Michael Lively[1], Brandon J. Holybee[1], Michael Toriyama[1], Scott Norris[2], and Stefan Facsko[3]

[1]University of Illinois at Urbana-Champaign
[2]Southern Methodist University
[3]HZDR (Helmholtz-Zentrum Dresden-Rossendorf), Institute of Ion Beam Physics and Materials Research

FIGURE 1: Upper Left: Side-view cutaway snapshot of the initial (i.e. early-stage irradiated by 500 eV Kr ions) surface used for the simulations. Lower Left: Side-view cutaway snapshot of the "control" surface after a ~4 ns simulated relaxation period. Right: Time-evolution of the Ga-Ga and Sb-Sb pair distribution functions, showing that Ga remains in an amorphous state while Sb self-arranges into nanocrystalline lattices.



EXECUTIVE SUMMARY

Directed irradiation synthesis of ordered nanodots and other nanostructures on III-V semiconductors has tremendous potential as a method for rational design of advanced nanomaterials for various industrial applications. Massive-scale atomistic simulations used while developing this technique are necessary to address knowledge gaps in existing theories of nanopattern formation and growth. Molecular dynamics studies have been carried out on Blue Waters to simulate the ion bombardment up to 8.4×10^{13} cm^{-2} fluence of a 100×100 nm^2 gallium antimonide (GaSb) surface of altered compositional state resembling experimental observations at the onset of pattern formation. The dominant observation is the formation of Sb nanocrystals in Sb-enriched GaSb regions leading to disparate mobilities of Ga versus Sb. The Sb crystallization is not directly ion-driven during the simulated time but is an intrinsic material response to the existing ion-induced compositional instability. The direct ion-induced mechanisms driving this instability remain the subject of additional, detailed study.

INTRODUCTION

The use of plasma- and ion-based synthesis has gained attention as scalable techniques to create a variety of surfaces using a single process step have been developed. The interfaces between plasma and material become an open thermodynamic system driven from equilibrium by physical mechanisms including high-energy kinetic disordering, compositional phase dynamics, and the emergence of metastable material states. The instabilities that arise lead to the evolution of well-ordered nanostructures, the compositional and morphological characteristics of which dictate the material properties.

Of particular interest is the formation of hexagonally-ordered quantum dots at the surfaces of III-V semiconductors such as GaSb exposed to energetic ion beams [1]. However, little progress has been made in modeling the evolution of III-V surfaces under ion irradiation, and thus the driving mechanisms behind pattern formation are poorly understood—in particular how compositional gradients near the surface of multi-component materials drive nanopatterning. Consequently, the dependence of the nanopattern on experimental parameters such as the incident ion energy or mass is understood in a broad, qualitative sense, making it difficult to design surfaces in regimes which have not been explored. Lacking is a comprehensive, atomistic computational modeling paradigm which addresses these questions of structural and compositional dynamics, coupled with directed in-operando experimental validation tools and techniques.

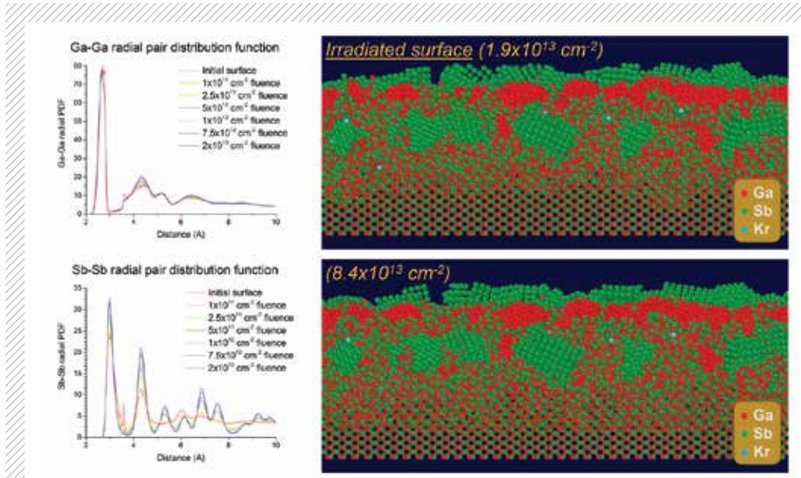


FIGURE 2: Left: Time-evolution of the Ga-Ga and Sb-Sb pair distribution functions for the irradiated surface, most importantly indicating that the self-ordering of Sb atoms is not directly influenced by the incident ions, by comparison to the same data in Figure 1. Right: Side-view cutaway snapshots of the surface after simulated ion bombardment to fluences of 1.9×10^{13} cm^{-2} and 8.4×10^{13} cm^{-2}.

Another challenge is identifying physical scales where experiments and models can be compared. Ion-induced mechanisms are thought to be ultrafast events that could be coupled to atomistic modeling at simulated time scales of the order of a few nanoseconds. Additionally, the ability to fabricate III-V nanodot arrays rapidly and reliably is critical for industrial applications. The understanding gained from this work lays a foundation for the development of advanced nanomaterials and surfaces to be used for devices in fields as diverse as semiconductors, biomaterials and biosensors, or next-generation energy technology.

METHODS & RESULTS

Simulations are started from an initial surface created with a compositional profile mimicking the early-stage ion-irradiated surface at the onset of patterning, which has been measured experimentally (surface representative of 500 eV Kr^+ irradiation to a fluence of nearly 10^{16} cm^{-2}). This technique allows detailed study of the mechanisms active during the actual pattern formation process, elucidating the influence of the surface composition on the ion-induced momentum and energy transfer and the presence of compositionally-driven surface responses which are not directly ion-induced. This initial surface is shown in the upper left of Figure 1.

In addition to the ion-bombardment simulations, a shorter "control" study was performed to identify compositionally-driven mechanisms which were not influenced by the ion impacts. For this study, the surface was allowed to run without ion impacts for ~4 ns. The results are shown in Figure 1. The Sb clusters in Sb-enriched regions self-arrange into nanocrystalline lattices, while the Ga clusters in Ga-enriched regions remain amorphous. While initially surprising, this result could be expected based on previous experimental observations [2,3]. This phenomenon suggests that the amorphous Ga atoms retain a much higher mobility than the Sb atoms which are strongly bound into rigid, crystalline structures. Indeed, this conjecture also agrees with experimental observations that Ga has orders-of-magnitude higher diffusivity than Sb in GaSb [4,5].

Results from the ion-bombardment simulations are shown in Figure 2. Qualitatively, the surface looks very similar to the "control" surface, and the coordination analysis indicates no change in the dominant Sb-crystallization phenomenon. This observation confirms that the formation of Sb nanocrystals in the sub-surface is not directly ion-driven during the simulated time, but is an intrinsic response of GaSb to the already-present compositional instability. The initial ion impacts induce mixing and redistribution of the near-surface

atoms, leading to a metastable compositional depth profile which includes the Sb-enriched region. The atoms in this region phase-separate laterally into pure Sb clusters and 50-50 GaSb, the former of which crystallize as observed here, forming the "seeds" which will drive the formation of nanopatterns on the surface.

Further analysis is needed to investigate the influence of compositional variations on the direct ion-impact energy and momentum transfer to the surface atoms. Such information is critical for determining the mechanisms and parameters relevant to generalized models of surface nanopatterning [6,7], allowing the prediction of pattern parameters and surface properties as functions of experimental parameters such as ion species, incident energy, or choice of surface component materials.

WHY BLUE WATERS

The work completed involved conducting MD simulations of 3 million atoms for nearly 50 million time steps. On a conventional supercomputing cluster this would take multiple years to complete. Large output data files were written at a fairly high frequency, necessitating a high-performance file system to minimize the impact on the overall simulation time. Production runs on Blue Waters were able to simulate up to 10 million time steps on 128 nodes in just 48 hours, allowing the majority of the production runs to be completed in less than two months actual time.

SIMULATING STRONGLY CORRELATED SYSTEMS: FROM FRUSTRATED MAGNETS TO MANY-BODY LOCALIZATION

Allocation: Blue Waters Professor/250 Knh
PI: Bryan Clark[1]
Collaborators: Hitesh Changlani[1], Eduardo Fradkin[1], Krishna Kumar[2], David Luitz[1], and Xiongjie Yu[1]

[1]University of Illinois at Urbana-Champaign
[2]Tufts University

EXECUTIVE SUMMARY

The key problem in condensed matter physics is connecting microscopic details to emergent phenomena. My group has used Blue Waters to develop this connection. We have focused on three main emergent phenomena: many-body localization, spin liquids, and superconducting systems. We describe the former two in this report.

Frustrated magnets arise from geometrically frustrated lattices, such as the Kagome lattice. We find strong numerical evidence for a chiral spin liquid in a natural spin system on the Kagome lattice under a magnetic field. Many-body localization is a phenomenon where statistical mechanics breaks down and quantum mechanics survives at infinite temperature. Using a **newly developed** method we show the saturation of entanglement for many-body localized states.

INTRODUCTION

Frustrated Magnets

In many cases, the electrons in a material are localized in space. The important physics can be captured by viewing the electrons simply as spins which decorate a geometric lattice and want to anti-align. On a square lattice there is a simple pattern which satisfies all electrons; every other electron points up (respectively down). In contrast, when the electrons are on a triangular lattice, this is no longer the case. Instead, while two electrons can be anti-aligned, the third electron must be aligned with one of the other two, causing geometric frustration. One of the most frustrated lattices is the Kagome lattice, which consists of corner sharing triangles. This inherent frustration makes it possible for exotic phases to arise. One such phase is the chiral spin liquid.

Many-Body Localization

Many-body localization is a phenomenon where statistical mechanics [1,2] breaks down and quantum mechanics manifests itself at finite (even infinite) temperature. This is particularly surprising as quantum phenomena are typically only prevalent at zero temperature. From a technological point of view, this phase is important because it may allow for the construction of a quantum computer and quantum memory that is robust to thermal noise.

METHODS & RESULTS

Frustrated Magnetism

Our approach to understanding electrons on the Kagome lattice was to perform an exact quantum simulation (using exact diagonalization) for the largest system size possible given our computational constraints. We simulate a system of spins with the nearest neighbor XY interactions under a magnetic field. Our computational ability allows us to simulate 48 spins requiring finding the lowest eigenvector of hundreds of matrices each of which are of size 377 million by 377 million. In doing so, we find ample numerical evidence that this system forms a chiral spin liquid.

The strongest evidence for the presence of a chiral spin liquid is as follows: A normal state of matter is insensitive to the topology on which the system lives. A magnet on a sphere, a donut, or a cylinder looks roughly the same. Topological states of matter such as spin liquids know the topology of their system and can "feel" the effect of distant boundaries. In our simulations, we slowly twist the boundaries of the system so that the spins interact differently across them. Once we make a full twist, the geometry of the system returns to where it started and amazingly enough, the system does not. Instead, the system picks up an additional quantized phase. The presence of this phase is a key signal of the topological phase of matter and shows that our system is a chiral spin liquid.

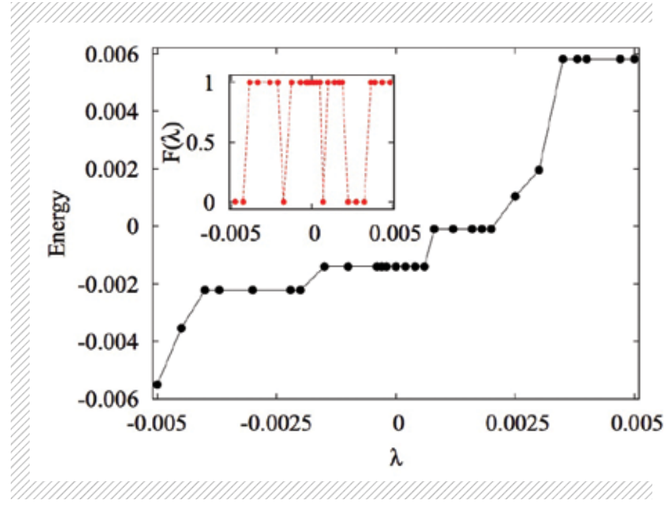


FIGURE 1: Example of sweeping the target energy lambda of a SIMPS run. Each dot corresponds to the eigenstate energy identified. The inset shows the overlap of nearest neighbor eigenstates. The fact that they are always one or zero validates that each state is individually an eigenstate.

Spin liquids are important for two reasons. First, they are the prototype for a phase of matter which stretches beyond the theoretical boundaries of the typical way physicists describe phases—the Landau paradigm. Secondly, they have the potential to be essential for the implementation of quantum computers that are robust to errors.

Many-Body Localization

Unfortunately, in the field of many-body localization, exact methods are limited to 22 sites. More sites are needed to better understand the phase, so we have developed a **novel** density matrix renormalization group (DMRG) algorithm (called the Shift and Invert MPS, or SIMPS). We applied this new algorithm to the disordered Heisenberg model scaling to an order of magnitude more sites than previously possible. With this capability we were able to measure the probability of entanglement, show the saturation of entanglement, and generate thousands of local excitations.

WHY BLUE WATERS

Frustrated Magnetism

Blue Waters was essential to "numerically prove" the existence of our chiral spin-liquid. Because of symmetry constraints, 48 spins was the smallest number of spins for which one could reasonably