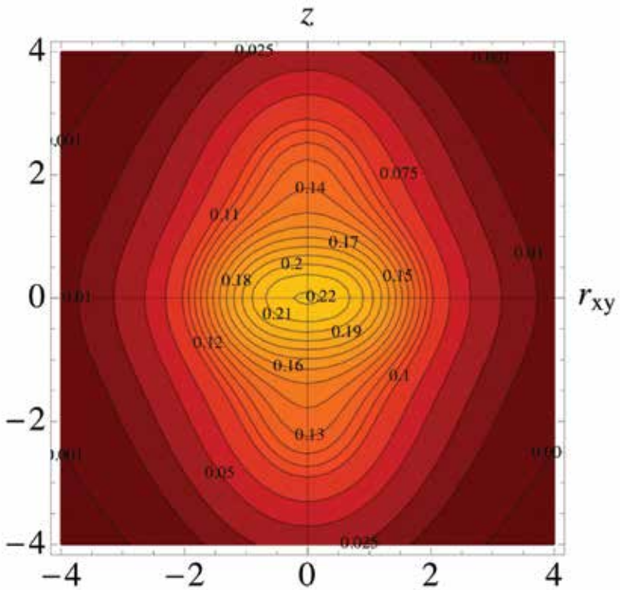

FIGURE 2: Density profile of the challenging ^{10}He ground-state resonance from first principles, revealing a surprising intrinsic structure.

matrix. The SA-NCSM drastically reduces the size of the problem and the associated memory requirement down to hundreds of terabytes and petabytes, but this comes at the cost of a major increase in computing intensity. As a result, SA-NCSM investigations of the intermediate-mass region are beyond the scale of available academic high-performance computing systems. Currently, only Blue Waters provides resources required for the *ab initio* SA-NCSM studies of medium-mass isotopes with cutting-edge accuracy. To capitalize on this opportunity, we drew from the experience and expertise of the Blue Waters staff and managed to improve the scalability of our code. As a result, our largest production runs utilized efficiently 717,600 concurrent threads running on **22,425 Cray XE6 compute nodes** to solve the nuclear eigenvalue problem with Hamiltonian matrices that occupy up to 400 TB of memory. Clearly, Blue Waters represents a unique computational platform that already plays a crucial role in advancing *ab initio* nuclear theory toward new domains.

NEXT GENERATION WORK

The major increase in computational power provided by the second generation of Track-1 system, along with emergent algorithms designed to take advantage of modern massively parallel architectures, will enable *ab initio* theories to start providing information of unprecedented quality for probing fundamental symmetries and physics beyond the standard model. Furthermore, increased computational resources will allow the SA-NCSM framework to address even heavier nuclear systems. For example, addressing neutrinoless double beta decay for ^{48}Ca should become feasible at a level that will reduce large uncertainties in the nuclear structure matrix elements and allows one to determine the neutrino type from planned experiments, which represents one of the most fundamental problems in physics today.

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NANOSCALE MECHANICS OF DEFORMATION IN HIGH-CAPACITY LITHIUM-ION BATTERIES

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EXECUTIVE SUMMARY

Silicon (Si) is one of the most promising electrode materials for high-performance lithium (Li) ion batteries because it has an order of magnitude higher specific capacity compared to conventional graphite electrodes. However, Si electrodes crack massively during Li insertion since they expand by 300% when fully-lithiated; they also delaminate from the current collector after many charge cycles. We have conducted large-scale parallel molecular dynamics simulations and density functional theory calculations on Blue Waters to uncover the underlying mechanisms for cracking and delamination of the Si electrode during charge cycling; these mechanisms are in excellent agreement with experiments. Blue Waters resources were needed because of the computational scale of the problem and the many computational runs needed to cover the entire parameter space. Our results have provided rich insights into the design and engineering of damage tolerant electrode materials for high-capacity Li-ion batteries.

INTRODUCTION

Lithium ion batteries are high-energy-dense systems that store energy by insertion of Li ions into solid electrodes. Silicon is one of the most promising electrode materials for high-performance Li-ion batteries since it possesses the highest known specific capacity of 4200 mAh/g, which is an order of magnitude greater than conventional graphite electrodes. During lithiation, the Si electrodes form Li_xSi compounds and undergo huge volume expansion of about 300% since one Si atom can theoretically bond with a maximum of x = 3.75 Li atoms. When attached to a metal current collector, such as copper (Cu), the massive and inhomogeneous

FIGURE 1: Sliding and delamination of Li_xSi thin film electrodes from the Cu current collector. (A) Atomic structure of the interdiffused Li-Si-Cu interphase between a Li_xSi electrode and a Cu-current collector. (B) Interface sliding facilitated by the formation of well-delineated and weakly bonded Si-Cu and Li-Cu crystalline atomic layers within this interphase structure. (C) Shear stress versus shear strain response demonstrating distinct regions of stress build-up and release leading to interface sliding, and stress accumulation leading to interface delamination.

volume changes during repeated lithiation and delithiation charge cycles lead to colossal cracking of the Si electrode. Recent studies have shown that Si electrodes of small feature sizes, such as nanowires, nanoparticles, porous structures, and thin films, display significantly higher reversible charge capacities and longer cycle life. In fact, a critical feature size of these nanostructured Si electrodes exists, below which fracture would be completely mitigated. It is believed that the improved fracture resistance originates from the ability of the nanoscale structure to accommodate the lithiation-induced strain by plastic deformation, resulting in lower stresses present during volume changes. However, the delamination of crack-free nanostructured Si electrodes from current collectors after a critical number of charge cycles has been widely reported, resulting in the loss of electrical contact and consequent capacity fade. Even though the cracking of Li_xSi thin films can be mitigated through patterning individual Si islands, the uncracked electrode still delaminates from the current collector after a critical number of charge cycles. To date, much is still unknown about the interface bonding the Si electrode and a metal current collector, such as Cu. Studies have suggested that sliding readily occurs along the Si-Cu interface to accommodate the massive volume changes in lithiated-Si during charge cycling. However, understanding the mechanisms of interface sliding and delamination is complicated by significant intermixing of Cu, Si, and Li atoms at the interface between a lithiated-Si film and the Cu substrate.

METHODS & RESULTS

Using first-principle calculations, we recreate model structures of the interdiffused Li-Si-Cu interphase (Fig. 1A) and show that the interdiffusion among Li, Si, and Cu atoms leads to the formation of well-delineated, crystalline Si–Cu and Li–Cu atomic layers at intermediate Li concentrations (Fig. 1B). These atomic layers are weakly bonded in shear, and readily slide to relieve the interfacial stresses during lithiation processes. Ideally, interface sliding between the Si electrode and the Cu current collector will help limit film stresses introduced by the lithiation process. However, sliding between the Si–Cu and Li–Cu atomic layers cannot occur indefinitely. The formation of pinning defects in the form of $LiSi_3$ compounds along the interface can eventually inhibit sliding (Fig. 1C). The consequential buildup of interfacial stresses leads to delamination failure of the Si electrode from the Cu-current collector. Understanding the atomic-scale mechanisms that promote or impede sliding provides the critical first steps toward designing Si-Cu interface structures to mitigate electrode failure.

WHY BLUE WATERS

To our knowledge, detailed analyses of the sliding and delamination processes of lithiated-Si electrodes from the current collector are virtually non-existent due to the computational complexity of such systems. Firstly, the lithiated-Si structures are amorphous. Recreating these structures requires substantial intermixing between Li and Si atoms through a heating and quenching process in *ab initio* molecular dynamics (MD) simulations and density functional theory (DFT) calculations which are computationally expensive. The computational complexity, coupled with a large number of computational runs to elucidate the interface mechanics as a function of lithium concentration, makes this a process that requires the capacity of Blue Waters. The proposed DFT and *ab initio* MD calculations were performed on Vienna *Ab initio* Simulation Package (VASP).

NEXT GENERATION WORK

Our next focus will be on elucidating the deformation and fracture mechanics of the Solid Electrolyte Interphase, which is critical to the performance of high-capacity lithium ion battery electrodes.

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LATTICE QCD ON BLUE WATERS

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EXECUTIVE SUMMARY

The goal of this project is to develop highly optimized code for the study of quantum chromodynamics (QCD) on Blue Waters to carry out calculations that will have a major impact on high-energy and nuclear physics. We have optimized and used the Chroma code for the simulation of Clover quarks and the MILC code for the simulation of HISQ quarks. Our long-term objectives with highly improved staggered quark (HISQ) are to generate gauge configurations with physical-mass up, down, strange and charm quarks, to use these configurations to calculate fundamental parameters of the standard model of high energy physics, and to perform precise tests of the standard model. The objective of our Clover quark program is to determine the excited mass spectrum of strongly interacting particles (hadrons) within QCD.

INTRODUCTION

The standard model of high energy physics encompasses our current knowledge of the fundamental interactions of nature. The model has successfully explained a wealth of data from accelerator and cosmic ray experiments over the last 40 years. However, it has been difficult to extract many of the most interesting predictions of quantum chromodynamics (QCD), the component of the standard model that describes the strong interactions. The only way to do so, from first principles and with controlled errors, is through large-scale numerical simulations. These simulations are needed to obtain a quantitative understanding of the physical phenomena controlled by the strong interactions, to determine some of the fundamental parameters of the standard model, and to test them. Despite the successes of the standard model, high-