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TOPOLOGIES OF ELECTRONIC STATES OF NANOCRYSTALLINE MATERIALS

Allocation: Illinois/50 Knh
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EXECUTIVE SUMMARY

The surface electronic states of crystalline solids determine a number of their physical properties. An exemplary class of materials for which surface states are intrinsically tied to a defining characteristic is the topological insulator (TI) family. The resistanceless surface conduction of TIs, which has potential applications in spintronic devices and dissipation-less transistors, originates from spin-protected surface electronic states. Topologically protected electron conduction in bulk TI materials has been the focus of many recent experimental and theoretical studies. However, the potentially diverse characteristics and topologies that these states may exhibit on the nanoscale have not been fully explored. Critical questions remain such as whether topologically protected surface states can exist on the surfaces of small nanocrystals. Blue Waters permits us to answer such questions through computations of realistic models of nanostructures consisting of thousands of atoms. Our computations will drive discoveries of novel semiconductor phases and electronic topologies in nanostructures.

RESEARCH CHALLENGE

Engineered nanocrystals have promise in new functional electronic and optical materials. Our laboratory makes use of unconventional techniques of nanomaterials synthesis that often produce novel compositions and crystal phases that are not found in the bulk phase diagram. Computational electronic structure investigations are allowing us to explore the properties of these unconventional nanomaterials and to discover novel physicochemical phenomena and principles.

As one key example, it is thought that novel electronic topologies may arise on the surfaces of nanocrystalline structures. The surface electronic states can determine key physical properties of a crystalline solid. A class of materials for which surface states are intrinsically tied to a defining characteristic is the topological insulator (TI) family. The resistanceless surface conduction of TIs results from the presence of spin-protected helical electronic states at the surface. However, questions remain as to whether topologically protected surface states can exist on the surfaces of small nanocrystals. If not, are there novel topologies associated with surface electronic states of nanocrystals? Blue Waters is a crucial resource that is permitting us to answer these questions. Our investigations may identify nanostructures with unconventional electronic transport properties of utility in energy storage and information technology.

METHODS & CODES

We use the open source Quantum Espresso software suite [1] to run electronic structure calculations using the plane-wave density functional theory (DFT) method. This software was deployed on Blue Waters as part of the work conducted with our prior exploratory allocation. We make use of Quantum Espresso's numerous parallelization levels that distribute large matrices and linear algebra operations among groups of nodes with good load-balancing. These parallelization schemes involve sizable and frequent communication among nodes, which relies on the speed of the Blue Waters communications hardware.

As our model system, we chose the $Hg_xCd_{1-x}Se$ alloy, where our calculations have predicted the presence of an inverted band structure and a nontrivial band gap (Fig. 1), two key attributes of a TI. In this model system, we are exploring whether there are critical differences in the nature of surface states of $Hg_xCd_{1-x}Se$ bulk slabs versus those of nanocrystals of $Hg_xCd_{1-x}Se$. What are the resulting implications for the electronic properties of the material? How do the surface states of $Hg_xCd_{1-x}Se$ nanocrystals change as the nanocrystal size is varied from 2 nm to 10 nm? We are interested in regimes where the nanocrystal electronic structure deviates from that of the bulk due to small-size effects.

Using DFT, we calculate electronic surface states of $Hg_xCd_{1-x}Se$ bulk slabs. Band-structure calculations coupled with projection of the electron spin permit the protected states of slabs to be identified. Study of size effects requires a different approach. Since nanocrystals below a certain size do not have bands, band-structure calculations cannot be used. We instead use the projected density of states and charge density maps obtained from calculations of whole nanocrystals. The projected density of states will reveal whether size regimes exist wherein quantum confinement enhances the band gap while retaining the protected surface states.

RESULTS & IMPACT

From our band-structure calculations, we are interested in uncovering how nanocrystal electronic structure and topology deviates from that of the bulk due to small-size effects. These deviations may amount to a complete disruption of the TI surface states. Alternatively, it might be found that nanocrystals of the appropriate size can retain the TI properties. From the electronic structure of nanocrystals that do or do not retain TI states, we will elucidate the underlying physics. This will allow us and others to tune electronic topologies by nanostructure design.

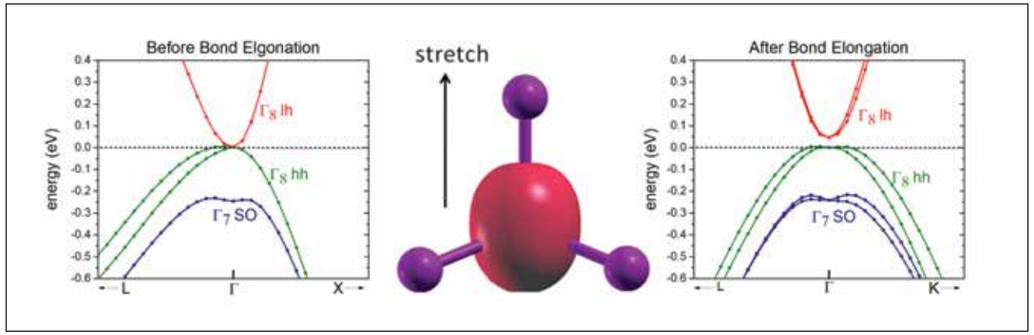

Figure 1: Our calculated band structures [3] show that in the novel wurtzite form of HgSe, symmetry breaking through elongation of Hg–Se bonds results in the opening of a band gap (right), whereas the conventional form of HgSe has no band gap (left). Hg atoms are shown in purple and the electron density of the highest energy valence band is shown in red (center).

The proposed studies will have marked impact on both the scientific community and society at large. Computational results demonstrating TI behavior in nanocrystals will put forth a new class of materials for study of fundamental physics and device applications [2]. Moreover, an understanding of the surface state characteristics will provide guiding principles to expedite new discoveries about unique electronic behavior on the nanoscale.

TIs are proposed to lead to energy-efficient logic devices because they provide a means to conduct electrons with almost zero resistance. This property could be used to reduce the amount of wasteful heat generated by logic devices, lowering the energetic cost of cooling and therefore enabling further scaling up of high-performance computing resources beyond current limitations imposed by overheating.

WHY BLUE WATERS

Our project requires use of the Blue Waters resource due to the computational scale of the bulk slab and nanocrystal calculations. Tens of nodes are required to fulfill the memory requirements of these calculations, exceeding the ability of small departmental clusters. Moreover, our investigation of the alloying and size effects requires not one but numerous such calculations. The computational expense of our effort would be prohibitive were it not for a Blue Waters allocation. Blue Waters will be able to accommodate any necessary increase in the size of our models. Furthermore, the specialized hardware of Blue Waters allows the Quantum Espresso code to run even more efficiently. Quantum Espresso's parallelization schemes involve sizable and frequent communication among nodes, which rely on the speed of the Blue Waters communication hardware.