THEORETICAL SPECTROSCOPY FOR TRANSPARENT CONDUCTING OXIDES

Allocation: Blue Waters Professor/0.245 Mnh
PI: André Schleife[1]
Collaborator: Joel Varley[2]

[1]University of Illinois at Urbana–Champaign
[2]Lawrence Livermore National Laboratory

EXECUTIVE SUMMARY:

Understanding the influence of dielectric screening on the electron-hole interaction and the resulting excitonic effects is a long-standing problem in computational materials science. Here, we achieve this for two transparent conducting oxides: In_2O_3 and Ga_2O_3. Both materials are very interesting in the context of transparent electronics and the semiconductor industry since they conduct electrical current while transparent. The unique combination of high-performance CPUs, large-memory configurations, and a large and fast storage system in Blue Waters allowed us to gain unprecedented insight into the physics of excitons and its impact on optical properties of these materials. We computed the most accurate theoretical optical spectra currently available for both materials and will use our results to further develop the computational framework to facilitate sustainable computational materials science and highly accurate theoretical spectroscopy for modern, complex materials that drive societal progress.

INTRODUCTION

Materials that are transparent and conducting are highly desirable for transparent electronics, photovoltaics, and optoelectronics. In this work we investigated indium oxide, In_2O_3, and gallium oxide, Ga_2O_3, since they are wide-bandgap semiconductors that show a remarkable combination of material properties. They are transparent across the visible spectrum and their electrical conductivity can be controlled over a large range, which enables a variety of novel device applications. In addition, the constituent elements are environmentally benign. Because of their properties, these oxides (amongst others) have been widely adopted as transparent conducting layers and they attract attention as stand-alone semiconductors in solar-blind photodiodes and Schottky diodes (e.g., for high-power electronics). Despite their appeal and widespread use, the influence of the electronic screening on the electron-hole interaction and the optical absorption is particularly poorly understood. This is difficult to measure directly in experiment, and the large unit cell size of In_2O_3 (bixbyite) and β-Ga_2O_3 (monoclinic) with up to 40 atoms has hampered theoretical studies, making the use of HPC necessary. Answering this question is not only important for device applications and the semiconductor industry, but it also provides insight for ongoing development of the computational framework itself.

METHODS & RESULTS

Our research is based on cutting-edge first-principles calculations within many-body perturbation theory. We computed optical absorption spectra using the Bethe–Salpeter equation framework based on single-particle energies obtained from a hybrid functional to approximate exchange and correlation. The bandgap from a hybrid-functional calculation was reproduced by a semi-local approximation to exchange and correlation and a scissor shift. This approach can meet the challenging convergence criteria necessary for optical absorption spectra.

Our theoretical spectroscopy results constitute the most accurate theoretical optical spectra available for these materials. Excitonic effects were explicitly included via the Bethe–Salpeter approach and for the entire spectral range studied here we found a pronounced influence of excitons. The strength of excitonic effects as a function of the photon energy will be explored in a follow-up project where we will extend our results to higher energies to compare with highly accurate experiments up to 40 eV. Our present work also advanced the qualitative understanding of optical anisotropy in monoclinic β-Ga_2O_3 both near the absorption edge and at high photon energies. This understanding is necessary for applications of these materials as transparent conducting oxides. From our present and future work in this context we learn how to model optical properties of technologically relevant materials (with societally important applications in photovoltaics, transparent electronics, and plasmonics) with very high accuracy using efficient parallelization and HPC.

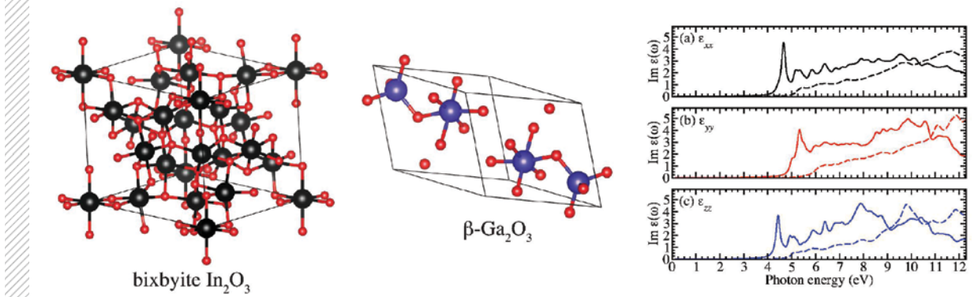

FIGURE 1: *Left and center:* Unit cells of In_2O_3 and Ga_2O_3 (In atoms: black, oxygen atoms: red, Ga atoms: blue) used in the present work. *Right:* Imaginary part of the anisotropic dielectric function (related to probability of photon absorption) of Ga_2O_3 (solid curves: excitonic and local-field effects included; dashed curves are computed without those) [1].

WHY BLUE WATERS?

For the present project, it was necessary to compute and analyze very large exciton Hamiltonian matrices. To study the optical properties of a single material in this work, we computed multiple matrices with ranks between 350k and 378k, leading to memory and storage demands between 0.5 TB and 1 TB for each individual matrix. Computing the matrix is a computationally demanding task that we accomplished using a well-parallelized Bethe–Salpeter code. The code used an ensemble run to divide the work into about 40 to 60 24-hour single-node jobs, each of which was parallelized across all cores of a given node using OpenMP. In this phase of the work, the large and fast storage capabilities of Blue Waters were important: Once the matrix was written to disk we used a time-propagation approach that scales quadratically with the rank of the matrix to compute an optical absorption spectrum. This code was parallelized using MPI; it read the matrix and distributed it into the combined memory of 16 Blue Waters nodes in less than five hours (62 MB/sec on average for the duration of 4.7 hours). The time-propagation scheme was then used to compute the optical absorption spectrum in about 6 hours.

The scenario described above was not a "one-shot" calculation. Our research requires multiple runs for each individual material and, hence, we needed a machine like Blue Waters that allows us to routinely carry out this work for multiple materials and configurations. The staff helped us with runs that needed more than the standard 24-hour wall time. Massive parallelism was not important to this work, but the entire "Blue Waters package" was. We needed high-performance CPUs that were very well integrated into large-memory nodes and connected to an excellent (large and fast) storage system. Blue Waters offered exactly this, along with outstanding availability, quick turnaround times, and a fast and very responsive support team.

We have already begun to extend this work towards much larger nanoscale systems. Those will require dealing with significantly larger matrices that lead to much more demanding requirements for memory, storage, and communication systems. For sustainable computational materials science that benefits society and, specifically, for predictive theoretical-spectroscopy techniques needed for computational design of nanoparticles for bioimaging or charge-separation in 2D materials, the next Track-1 system will be crucial.

PUBLICATIONS

Varley, J. B., and A. Schleife, Bethe–Salpeter calculation of optical-absorption spectra of In_2O_3 and Ga_2O_3. *Semicond. Sci. Tech.*, 30 (2015), 024010, doi:10.1088/0268-1242/30/2/024010.