Research accomplished in 2020:

In the first 10 months of 2020 we used Blue Waters for different projects in my group, related to electronic, optical, and magnetic properties of materials. We performed first-principles simulations of total energies, using density functional theory, to study the energetics of different Neel vector alignments in the metallic antiferromagnet Fe2As. From these simulations, we extracted the magnetocrystalline anisotropy and compared to experimental data obtained in the Illinois MRSEC. In addition, we predicted also the out-of-plane anisotropy, which is experimentally not easily accessible. These results are published in Physical Review B.

In addition, we performed first-principles simulations of single-atom point defects in transition-metal dichalcogenides. We introduced point defects into relaxed geometries of WSe2 and converged the resulting structural rearrangement with respect to simulation cell size. From our data we were able to extract the dependence of the relaxation on the distance from the point defect and compared the resulting pattern to experimental results from Pinshane Huang's group. This comparison helped explain an experimentally observed oscillatory behavior. These results are published in Nano Letters.

We also studied pre-equilibrium stopping power and projectile charge capture in thin aluminum sheets irradiated by 6 - 60 keV protons. Our time-dependent density functional theory calculations reveal enhanced stopping power compared to bulk aluminum, particularly near the entrance layers. We propose the additional excitation channel of surface plasma oscillations as the most plausible explanation for this behavior. We also introduce a technique to compute the orbital-resolved charge state of a proton projectile after transmission through the sheet. Our results provide insight into the dynamics of orbital occupations after the projectile exits the aluminum sheet and have important implications for advancing radiation hardness and focused-ion beam techniques, especially for few-layer materials. These results are also published in Physical Review B.

Finally, we used Blue Waters for various development work around our time-dependent DFT code and implemented a laser field (time-dependent electric field) into the code. Simulations that explore this implementation further and apply it to both MgO as well as a slab of aluminum are currently underway. From comparing these simulations to our results for particle irradiation in both systems we expect to understand the creation of such electronic excitations better and, potentially, connect to experiments in the future.

Research planned for 2021:

In Spring of 2021, PI Schleife will be on sabbatical and will use this time to further explore the connection of electronic excitations and emerging non-adiabatic dynamics. To this end he will explore laser irradiation of defects in materials as a means to enhance their diffusion and also the existence of defects as a reason for light-induced degradation. Depending on COVID this sabbatical will be either in Santa Barbara or in Germany (or both) and the respective set of collaborators will determine the specific materials to connect to. Such simulations can be important when defects are to be introduced into materials, e.g. for Qbits. This part of the work will also include implementation of absorbing boundary conditions into the code. Any newly developed code will be made available publicly via Github.

Other research planned for the 2021 period includes optical properties of semiconducting nanorods, which is work we are performing together with the experimental group of Prof. Smith in Bioengineering at UIUC. This will entail structural relaxations of large numbers of atoms, surrounded by vacuum, and subsequent simulation of single-particle optical spectra from density functional theory. We are also aiming to understand the influence of spin-orbit coupling on optical properties of CuI. The ultimate goal

of these simulations will also be to determine the limitations of time-dependent DFT in describing electronic relaxation and to explore other approaches, such as solution of the Boltzmann transport equation to overcome/circumvent these limitations.

Together with our collaborators in the Illinois MRSEC we are envisioning to simulate temperature-dependent optical spectra of metallic antiferromagnets. This will enable us to understand the influence of disorder in the magnetic subsystem on the optical and magneto-optical properties of materials.

Resources requested: We request to use 240k node hours and the default storage allocation.
Estimated use: Q1: 30 %, Q2: 35 %, Q3: 30 %, Q4: 5%

Data migration plan: Input and important output files of all simulations will be retained and we will migrate all data off Blue Waters either to our local storage device, which stores up to 20 TB, or to the Materials Data Facility, using the Globus interface. This process will begin in the last quarter of 2020, to migrate all files off the Nearline storage and will extend across 2021 for newly generated files. Not all data will be retained and this will be decided based on file size and computational cost to regenerate certain data.