

Project Title: Exploring novel Green's function-based *ab initio* methods for the prediction of optical properties of molecules.

Name of Applicant: Dr. Andrey Semichaevsky

Rank: Associate Professor

Department: Chemistry & Physics

Total amount of funding requested: \$6000

Salary supplements for projects requiring summer work: \$6000

Date of the application: April 10, 2021

Project outline

First principles or *ab initio* calculations [1] are commonly used in computational physics and chemistry to predict distributions of electron density and the electronic structure of molecules, clusters of atoms, and of continuous periodic lattices in solids.

Traditional mean-field theory methods, such as density functional theory (DFT) [1] are successfully applied to the analysis of ground-state band structures of solids and electronic states of molecules in gases, mechanical properties of solids, calculations of phonon spectra.

There exist, however, fundamental issues that result in major limitations and require precautions when one uses mean-field approximations in the analysis of excited electronic states and of optical properties [2]. Applying approximations based on one-particle Green's functions can lead to significant uncertainties, if one wants to find: quasiparticle interaction energies, the spectra of optical absorption and emission, fine energy structure, carrier lifetimes, effects of doping on material bandgap.

The applicant's interest in the subject began during his last year in the ECE doctoral program at UMass (2005-2006). Then he worked with a physics faculty, Dr. Kussow (now retired), who introduced him to advanced quantum mechanics. That project was funded by the AFRL and had a focus on optical metamaterials that exhibit negative effective refractive index over a band of frequencies. Preliminary work for the project was carried out using Siesta and ABINIT codes in 2006-2007, and its results were presented at a physics conference [3].

Recently, Dr. Kussow contacted the applicant with a novel idea about how the limitations of the mean-field approach in optical *ab initio* calculations can be overcome. In order to do that, one would have to solve a many-particle interaction problem using two-particle excitations with the corresponding Green's functions applied as tools. The draft of that paper was sent by the collaborator to famous journals like The Annals of Physics, Physical Review, and Physica Status Solidi, but was rejected by them because the original paper had no practical calculations or examples.

Therefore, in this proposed project, the applicant will use preliminary results to construct a model, write a practical code, and compute a couple of examples of excited state energies in simple gas molecules, for which experimental data on optical absorption spectra or ionization energies are readily available. This will help the applicant and his collaborator complete the manuscript and to submit it for publication.

References

1. Andrew Leach. Molecular modelling: Principles and applications. 2nd edition. Prentice-Hall, 2001.
2. Michael Rohlfing, Steven G. Louie, PHYSICAL REVIEW B. **62**, 8, 2000, 4927.
3. A. Akyurtlu, A-G. Kussow, A. Semichaevsky, "First-principles studies of the optical properties of MgB₂ and its applications in optics," URSI Annual Meeting, Ottawa, Canada, July 22-26, 2007.
4. P. Gianozzi, *et al.*, Journal of Physics: Condensed Matter, **21**, 39, 2009, 395502.

Measurable goals and objectives

The goal of this research is to develop a computational model for excited states in many-electron atoms. This will help other scientists improve predictions of optical properties of molecules and atomic clusters (nanoparticles), to include more details in existing models, and to gain better understanding of experiments, such as ionization of noble gases in intense laser beams.

The project has two objectives:

- 1) To develop and to test a code written in a language like C++ that would include: evaluation of generic 2-particle Green's functions, evaluation of optical excitations, electron density, and prediction of optical properties;
The code will be tested on gas molecules, using atomic pseudopotentials included in Quantum Espresso [4];
- 2) To finalize the manuscript, prepare and submit it to Physica Status Solidi (PSS) or a similar journal;

Time frame and resources.

The plan is to begin preparations for this project in June and complete it by August 2021. The total time required is about 160 hours, roughly 20 hours per week over 8 weeks, which is equivalent to 1 month of work, spread over 2 months. The applicant has enough computer resources to carry out this project.

How will the project enhance teaching and research at Lincoln University?

Main contribution of this project is expected to be a peer-reviewed publication and a computational tool that can be further used by the applicant and his collaborator in future scholarly work. Our department is trying to bring back upper-level students into our classes. We have just submitted a \$613,000 NSF S-STEM grant that would sponsor up to 10 undergraduate scholars yearly. The results obtained through this project may eventually find their way into undergraduate courses, including physics and chemistry ones. Among them are Quantum Mechanics, Computational Physics, Research Seminars, and Independent Studies.

How the success of the project will be measured?

One of the measures of success is a successful peer-reviewed paper or a conference paper. Another one is the prototype computer model that successfully computes excited state energies for simple gas molecules based on the new approach that we propose.

How, when, where, and with whom the project's outcome will be shared?

The results of this project will be presented at conferences outside Lincoln and in a departmental seminar series at Lincoln.

Budget

Description	Time frame	Rate	\$ amount
Development and testing of new <i>ab initio</i> code.	06.01.2021 to 07.31.2021	\$3000 per month	\$6000
Total	\$6000		

Note: the manuscript to be submitted is about 75% ready, and we also plan to use other funds, such as departmental allowances to publish it.

(To be used only by the Applicant)

Name of the Applicant: Dr. Andrey Semichaevsky

Rank of the Applicant : Associate Professor

Submission Date: 4/10/21

Expertise Area : Physics, electrical engineering.

Prior FD Proposal Outcomes: Development of lab-lectures for physics and engineering courses.

Proposal Submission Term: Please circle one of the following

1. Spring 2021
2. Summer 2021
3. Fall 2021

Faculty Development Committee (FDC) Proposal