# Press Release University of Bremen and University of Oldenburg

The Bremen Center for Computational Materials Science at University of Bremen and the Ultrafast Nanooptics Group of University of Oldenburg are happy to announce, **that Dr. Sergei Tretiak from Los Alamos National Laboratory has been honored with the Alexander von Humboldt Research Award**. Both, the host Prof. Dr. Thomas Frauenheim and Dr. Sergei Tretiak share **common research interests in modeling of photo-induced phenomena in extended molecular systems, condensed matter, nanoscale materials and interfaces, and devices**.

Thomas Frauenheim is a pioneer and expert in developing and applying *density functional tight binding* (DFTB) theory. On the other side Sergei Tretiak is well known for developing and applying a broad spectrum of methods, known under the rubric of *non-adiabatic excited state molecular dynamics* (NEXMD) and aimed at modeling non-equilibrium processes in nanoscale materials in the real time-domain at the atomistic level. The expertise of the nominator and nominee are complementary, and their collaboration will lead to significant breakthroughs in materials science and photo-induced non-equilibrium phenomena. In particular, DFTB allows one to treat systems that are of technological relevant sizes.

The recently developed collaboration between the nominator, the nominee and the spectroscopy group at the University of Oldenburg led by Prof. Dr. Lienau led to a ground-breaking scientific discovery about ultrafast charge and energy transport across intermolecular Conical Intersections (CoIns) in molecular aggregates.

These seem to control the flow of energy in light harvesting function of soft materials, a key process in photovoltaics. In this work, the NEXMD software was used to provide supporting theoretical modeling, and the nominee and the nominator jointly supervised the work of the postdoc and led efficient interaction with the experimental team of Christoph Lienau in Oldenburg. This case exemplifies expected interactions and research supported by the Humboldt Award, which is envisioned in the future. Specifically, together with the Lienau group in Oldenburg the collaborators will explore the role of intermolecular CoIns on the energy relaxation dynamics in thin films related to organic photovoltaic materials, aiming to intentionally control the dynamics of energy and charges in the process of light harvesting, which will be funded by a DFG project in the next three years.

New modeling requires an extension of the NEXMD capabilities to directly simulate the coherent two-dimensional electronic spectra (2DES) that are measured experimentally. Moreover, a new experimental idea is to invoke coupling to external resonator modes, exploring polaritonic phenomena. Modeling polaritonic chemistry requires incorporation of electronic, nuclear, and photonic degrees of freedom into the NEXMD simulations interfaced to the DFTB+ software. The Tretiak group at Los Alamos has already performed some preliminary studies of these novel modeling extensions, including advanced algorithms for non-adiabatic molecular dynamics simulations (e.g., Multiticonfigurational Ehrenfest Ab Initio Multiple Cloning, MCE-AIMC), interface of NEXMD with SPECTRON code to directly calculate 2DES, and implementation of non-adiabatic algorithms to account for strong light-matter interactions (i.e. polaritonic effects). Overall, the collaborating groups believe that tuning these coherent dynamics will provide new opportunities for steering the flow of energy and charges and their pathways on the nanoscale in functional assemblies for energy storage and may lead to new routes to the design of optoelectronic devices.

**More infromations:** The collaborating groups at the Universities of Oldenburg, Bremen and Los Alamos National Laboratory further intend to expand their immediate research projects

to a broader scope of *photoinduced chemical processes*, such as water splitting and carbon dioxide and nitric oxide reduction. The studies require models that include an inorganic semiconductor, acting as a catalyst, and a solvent. Such models should take into account multicomponent systems of practical interest such as $TiO_2$ nanocrystals, layered 2D hybrid perovskites, composites of carbon nanotubes/graphene flakes, ligated semiconductor nanocrystals, etc. DFTB can treat systems including thousands of atoms, greatly expanding the range of systems and processes that can be studied. The non-equilibrium processes involved in the photoinduced charge separation and transport require explicit *time domain modeling*. They occur on *ultrafast timescales* and frequently cannot be described by rate expressions. Charge separation, transport of electrons and holes, energy losses to heat due to charge-phonon interactions, energy transfer, and electron-hole recombination *occur in parallel and compete*. Treating all these phenomena presents an exciting theoretical challenge that will be addressed by our collaboration.