CFEL SCIENCE

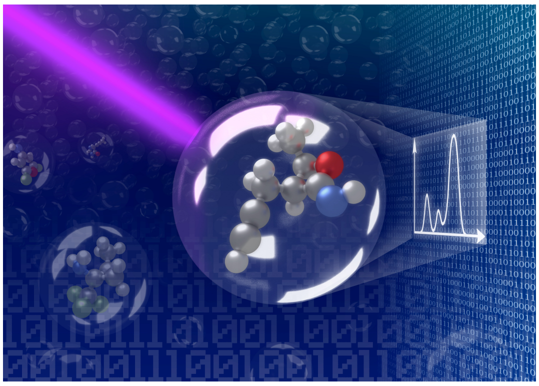
SEMINAR

18th January 2023 - 2:00 p.m.
CFEL-bldg. 99, seminar room IV

Patrick Rinke

Aalto University School of Science, Aalto, Finland

Recent developments in theoretical spectroscopy: core excitations, strong correlation and artificial intelligence



The *GW* and the Bethe-Salpeter (BSE) approximations to many-body perturbation theory (MBPT) have been incredibly successful in theoretical spectroscopy to model electron addition and removal and absorption spectra. Yet, challenges abound and new frontiers await. In this presentation, I will introduce our first exploration into core excitations. We have implemented the real-frequency contour deformation *GW* approach into the all-electron numerical atomic orbital code FHI-aims, added efficient relativity corrections for s-states and solve BSE for core states as measured in X-ray absorption (XAS). Our benchmarks reveal that computed core level binding energies deviate by generally less than 0.3 eV from experiment (on an absolute scale) outperforming the popular density-functional theory based Delta Self-Consistent Field (ΔSCF) method. Conversely, K-edge excitation energies agree within 0.6 eV with experiment.

To tackle strongly correlated systems, we have developed a new quantum embedding theory. It captures strong (static) correlation in a subspace by configuration interaction (CI) theory and high-energy dynamic correlation with MBPT in the *GW* and BSE approximation. For the challenging multi-reference problems of H_2 and N_2 dissociation, we obtain good agreement with benchmark results. Our theory treats ground and excited states on equal footing, and we compute vertical excitation energies of N_2 and free-base and Mg porphyrin in excellent agreement with high level quantum chemistry methods.

Despite the successes of theoretical spectroscopy, calculations and experiments are costly and time consuming. For this reason, we recently enhanced theoretical spectroscopy with artificial intelligence (AI). Once trained, the AI can make predictions of spectra instantly and at no further cost. I will present artificial neural networks that can learn excitation spectra of molecules with an accuracy of 97%.

Host: Robin Santra – CFEL-DESY Theory Division