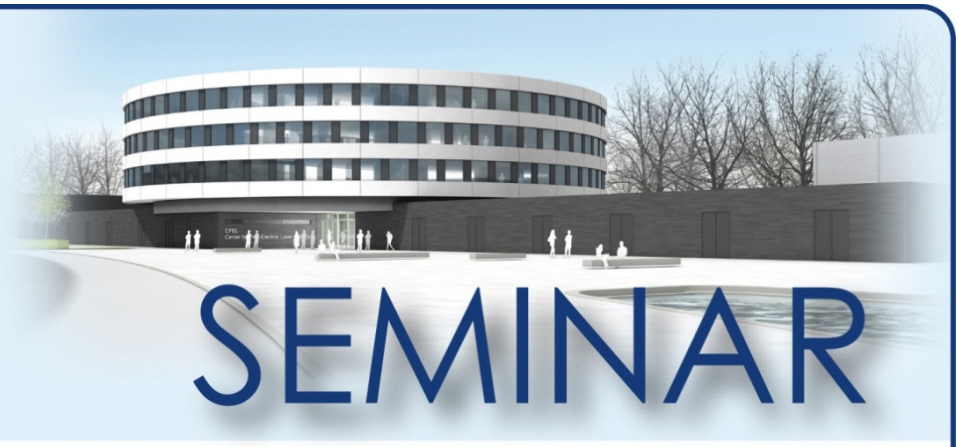
CFEL SCIENCE



SEMINAR

9th January 2019 - 2:00 p.m.

CFEL-bldg. 99, seminar room IV

Anna Krylov

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Theory and computational modeling of two-photon absorption processes: From quantitative predictions to qualitative insight

Non-linear optical (NLO) properties can be exploited in a variety of exciting applications. For example, two-photon absorption (2PA) spectroscopy enables 3D and super-resolution imaging, nanosurgery, high-density optical data storage, and targeted cell deactivation. The design of new materials and interpretation of 2PA spectra require fundamental understanding of the quantum-mechanical principles behind the NLO phenomena. This lecture will outline the theoretical foundations of 2PA phenomena, describe the formalism for calculating 2PA cross-sections within the equation-of-motion coupled-cluster theory, and introduce a framework for a rigorous mapping of the 2PA transitions between many-body states into a simple one-electron molecular orbital picture. The capabilities of the theory will be illustrated by examples (2PA spectra of prototypical conjugated molecules). The extension of the theory of the NLO phenomena into the X-ray regime will be discussed.

The following aspects will be emphasized:

- Strategies for accurate calculations of 2PA cross-section using correlated wave functions;
- Computational efficiency;
- Molecular orbital theory of 2PA processes and visualization of virtual states;
- Exact and approximate selection rules.

[1] A.I. Krylov, Ann. Rev. Phys. Chem. 59 433 (2008).
[2] K. Nanda and A.I. Krylov, J. Chem. Phys. 142 064118 (2015).
[3] K.D. Nanda and A.I. Krylov, J. Phys. Chem. Lett. 8 3256 (2017).

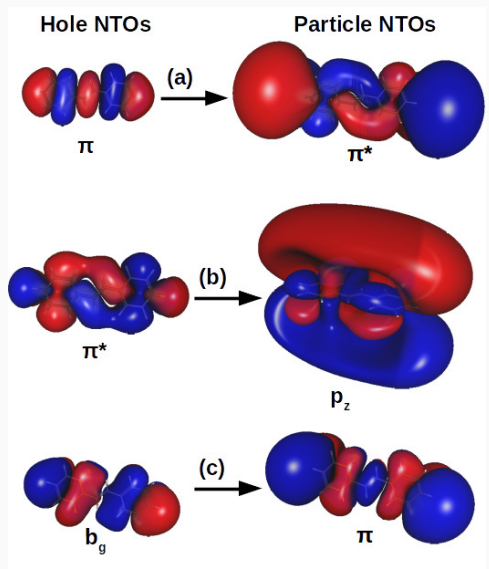

Figure 1. Natural transition orbitals for a bright 2PA transition ($1A_g \rightarrow 5A_g$) in trans-stilbene computed using EOM-CCSD/ daug-cc-pVDZ wave-functions.

Host: Robin Santra – CFEL-DESY Theory Division