Summary for general public

The fundamental aspects of light-matter interactions, especially in the strong-field limit, are still active area of research. One of the nonlinear optical processes, i.e. multiphoton absorption, is in limelight due to attractive characteristics with potential for applications in material sciences including data storage, and biological microscopy. By employing two- or three-photon-excited fluorescence probes in bioimaging one may limit the scattering effects, improve resolution and increase penetration depths within the tissue. In the multiphoton absorption process, the simultaneous absorption of two-, three- (and more) photons occurs thus allowing for reaching a higher excited state, provided that the resonance condition is met. The only well-studied multiphoton absorption process is two-photon absorption (2PA), while three-photon absoprtion (3PA) – central to the current project – has been studied scarcely. This holds for experimental as well as theoretical works. Over the last decades guidelines were developed to design efficient two-photon absorbers. Still, it has not been verified to what extent the existing rules are transferable to the three-photon absorption. **The primary goal of the current project is to study strategies for maximizing 2PA and 3PA efficiency on an equal footing in a systematic manner.** With bioimaging applications in mind, the development of three-photon absorbers as molecular probes for microscopy requires fulfilling additional requirements, e.g., tuning of absorption wavelengths and fluorescence quantum yields. Unfortunately, the design rules have not been set yet. **Moreover, in the present project we will make an attempt to fill this gap and develop design rules for three-photon-excited fluorescent probes**. To that end, a synthesis of novel fluorescent probes will be performed, followed by spectroscopic characterization (including determination of one-, two- and three-photon electronic spectra), theory development and advanced computer simulations. An important outcome of this project will be the comparison of the two- and three-photon absorption efficiency – determined in the very same experimental conditions – and interpretations of these results in terms of electronic structure theories for future use in molecular design. The joint experiment-theory efforts towards establishing structure-property relations for three-photon absorbers will be performed for fluorescent dyes belonging to the family of organoboron complexes.