Theoretical principles and computational design of two-photon absorbing photochromic chromophores

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Exponential growth in Information Technology generates ever increasing amounts of data. This made recording density of storage media crucially important. Photon-mode optical recording is expected to win the competition with magnetic and heat-mode optical recording. Two-photon absorption was proposed as a basis for high-density multi-layer technology for optical memory and logic devices. This technology is suggested to use polymers, doped with photochromic compounds that undergo a reversible photoinduced isomerization, or photoswitching. Our goal is to combine the photon-mode recording abilities of photochromic compounds with large two-photon absorption (2PA) cross-sections. Here we investigate a prototype molecule where a diphenylaminofluorene moiety with a large 2PA cross-section is covalently attached to an efficient photoswitch (diarylethene). This prototype was shown to be photochemically inactive in experiment. We analyse its electronic structure using Density Functional Theory, identify a nature of this inactivity and propose the general guidelines for the rational design of a 2PA photoswitch. We also compare our predictions with experimental 2PA spectra for polymethine chromophores in both solution and crystalline phase.