Two-photon absorption spectra of the spiropyran and merocyanine pair: A comparative study of solvation models

A. Roztoczyńska[1], R. Zaleśny[1], N. Arul Murugan[2], J. Kongsted[3], J. Olesiak-Bańska[1], K. Matczyszyn[1], W. Bartkowiak[1], M. Samoc[1], H. Ågren[2]

[1]Institute of Physical and Theoretical Chemistry, Wroclaw University of Technology, Poland

[2] Division of Theoretical Chemistry and Biology, Royal Institute of Technology, Sweden

[3] Department of Physics, Chemistry and Pharmacy, University of Southern Denmark, Denmark

E-mail: agnieszka.roztoczynska@pwr.wroc.pl (A.R.)

In their seminal paper, Parthenopoulos and Rentzepis demonstrated the potential application of a photochromic molecule embedded in a polymer matrix for three-dimensional optical storage memory [1]. The device employed two-photon writing and reading. Since then, many follow-up studies have been devoted to optical properties of other photochromic systems for numerous applications including optical data storage (see Ref. [2, 3, 4] and references therein). Computer-aided design of two-photon active media has proven its appreciable role in molecular nonlinear optics. In fact, first-principles quantum chemical calculations allow to ivestigate (electronic) structure-property relationships and their results are often stimulus for experimental studies. However, the reliability of the predictions heavily depends on the level of theoretical approximation. In the present study, we report the one- and two-photon absorption spectra of the spiropyran/merocyanine pair in aqueous and chloroform solutions. The spiropyran molecule in question exhibits photochromism both in solution and in the crystalline state [5]. In order to study the linear and nonlinear absorption spectra in solutions, we employ several computational strategies, with an eye towards their comparison. In particular, the set of studied approaches includes the polarizable continuum model and a newly developed polarizable embedding self-consistent scheme (linear in the solvent polarization) [6]. The results of the calculations are compared to experimental data obtained with the aid of the Z-scan technique.

The work was financed by a subsidy from the Polish Ministry of Science and Higher Education within "Iuventus Plus" programme (Grant No. 0628/IP3/2011/71).

[1] Parthenopoulos, D. A., Rentzepis, P. M. *Science*, 245:843-845, 1989.

[2] Irie, M. *Chem. Rev.*, 100:1685-1716, 2000.

[3] Yokoyama, Y. *Chem. Rev.*, 100:1717-1740, 2000.

[4] Berkovic, G., Krongauz, V., Weiss, V. *Chem. Rev.*, 100:1741-1754, 2000.

[5] Bénard, S., Yu, P. *Adv. Mater.*, 12:48-50, 2000.

[6] Olsen, J. M., Aidas, K., Kongsted, J. *J. Chem. Theory Comput.*, 6:3721-3734, 2010.