

MEASURING BROADBAND TWO-PHOTON ABSORPTION SPECTRA WITH ACCURATE ABSOLUTE CROSS-SECTIONS IN SOLUTION

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Two-photon absorption (2PA) spectroscopy measures the simultaneous absorption of two photons, where the combined energy of the two photons corresponds to an allowed transition of a molecule. 2PA is a powerful spectroscopic tool that can probe the electronic structure of a molecule by accessing different electronic states, than in one-photon absorption (1PA) spectroscopy because 1PA and 2PA are governed by different selection rules. Many applications rely on nonlinear two-photon excitation, including 3D fluorescence imaging and photo-dynamic therapy; however, broadband 2PA spectra and accurate absolute cross-sections are rarely reported in the literature. Our broadband 2PA measurements use a femtosecond pump-probe technique that is uniquely able to fill this need by measuring continuous 2PA spectra and accurate absolute 2PA cross-sections using stimulated Raman scattering (SRS) as an internal standard. Our broadband 2PA technique also recovers additional information based on the relative polarization between the two photons, giving insight into the symmetry of the excited electronic states of a molecule. We report 2PA spectra for coumarin 153 in several solvents, for Mn and Re coordination compounds and for pure liquid benzene.