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Two-Photon Optical **Properties** of AIEactive D-TPE-A Molecules: Aggregation Enhancement and Structure-**Property Relationships** YILIN ZHANG, Department of Physics, the Hong Kong University of Science and Technology, JIE LI, BEN ZHONG TANG, Department of Chemistry, the Hong Kong University of Science and Technology, KAM SING WONG, Department of Physics, the Hong Kong University of Science and Technology — We present an aggregation enhancement in two-photon-excited fluorescence (TPEF) of about two orders of magnitude in a series of novel non-centrosymmetric $D-\pi$ -A molecules. Aggregation-induced emission characteristics are introduced into these D- π -A molecules via tetraphenylethylene (TPE), which is used as their π bridge. Detailed analysis shows that the TPEF of these molecules are enhanced in aggregation environment with both fluorescence quantum efficiency and two-photon absorptivity concomitantly. The two-photon absorption (TPA) transition bands of these branched- or butterfly-configured molecules are similar to those in their linear absorption. The molecular TPA cross sections in aggregation environment reach around 50-130 GM, and peak within the available wavelength ranges of a Ti:Sapphire femtosecond oscillator. We also observe that two-photon absorptivity increases progressively with the addition of donor/acceptor moieties on the TPE backbone. This phenomenon is presumably attributed to the improved conjugation length and enhanced intramolecular charge transfer, hence better delocalization of π -electrons. For each compound, the aggregation enhancement in TPA may also offers clues of aggregation effect on the molecular electronic structure.

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