Corrigendum

Strong Two-Photon Excited Fluorescence and Stimulated Emission from an Organic Single Crystal of an Oligo(Phenylene Vinylene)

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In this Communication Figure 5 and Figure S5 in the Supporting Information were misinterpreted as a result of an experimental artifact. While we wrote on page 734, that two-photon excited fluorescence of TOPV in the solid state can be unambiguously recorded even on a standard spectrofluorimeter with a Xe lamp as excitation source, a critical reader noticed that a high-intensity laser is still needed to promote a material in the excited state by a two-photon absorption process. In additional experiments we found that the observed emission applying an excitation at 800 nm was a result of the higher order of the diffraction light in the monochromator of the spectrofluorimeter used. Thus, there are also photons available at 400 nm for one-photon excitation, although 800 nm was set as the excitation in the equipment. Diffraction light of a higher order in the monochromator caused the observed emission. This mistake happened as a result of relying on the specifications of the commercial equipment used for the experiments, which suggests that higher-order diffraction light from the monochromator is completely filtered out. This assumption led us to draw an incorrect conclusion because we believed our material has such a strong two-photon absorption that just a Xe lamp could be sufficient to promote the material by a two-photon absorption process. As the content that refers to Figure 5 and Figure S5 is based on a wrong assumption this part of the article is incorrect. A high-intensity laser source is still needed for two-photon excitation, as described elsewhere.[1]

Soon after the publication of our Communication, we noticed a new article by Spano[2] which revealed that some organic dyes that were formerly thought to form J-aggregates based on spectral shifts are actually H-like aggregates. Further investigation suggested that the TOPV molecules may exhibit similar behavior. Although comparison between the molecular and powder absorption (Figure S3 in the Supporting Information) suggests a red-shift of the absorption in the solid state versus in solution, TOPV molecules might form weakly coupled H-aggregate rather than J-aggregate as we had ascribed in our Communication. The small bump at 467 nm in the powder emission spectrum (Figure S3 in the Supporting Information) become apparent in nanoparticles with increasing molecular disorder, and therefore can be assigned to the 0–0 band of H-aggregates.[2,3]

The table of contents entry may also confuse some readers. The meaning of "Low in, high out" used in the Communication refers to the nature of TPA phenomenon. The TOPV molecule emits one higher-energy photon after absorbing two low-energy photons. No quantitative two-photon absorptivity of the crystal is meant in this context.

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[1] B. Strehmel, V. Strehmel in *Advances in Photochemistry*, Vol. *29* (Eds.: D. C. Neckers, W. S. Jenks, T. Wolff), Wiley, Hoboken, **2007**, pp. 111 – 341.

[2] F. C. Spano, *Acc. Chem. Res.* **2010**, *43*, 429.

[3] J. Gierschner, M. Ehni, H.-J. Egelhaaf, B. Milián Medina, D. Beljonne, H. Benmansour, G. C. Bazan, *J. Chem. Phys.* **2005**, *123*, 144914.