Supplementary Information to Comment on entangled two-photon absorption in molecules.

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The properties of entangled two-photon absorption (ETPA) are manifested in else number of experiments.

Several groups of physicists are studying ETPA in molecules. At the same time, some physicists observe a great superiority of ETPA over the classical two-photon absorption (up to six orders of magnitude in cross-section or radiation intensity) [1 - 5]. But other physicists in similar experiments do not observe such superiority at all [6 - 8]. In [9], such a difference in the results is explained by a possible uncontrollable difference in the initial conditions of the experiments.

Other groups of physicists are studying IR imaging through two-photon absorption [10 - 15]. The two-photon absorption (TPA) process here takes place directly in the semiconductor detector. It has been found that the so-called nondegenerate two-photon absorption (ND-TPA) is much more effective (by two to five orders of magnitude in radiation intensity) than the same degenerate two-photon absorption (D-TPA). The authors have no any doubts about this superiority of ND-TPA over D-TPA. However, the authors also do not have a physical explanation for this superiority.

The physical explanation is that in the case of ND-TPA, the authors are dealing with entangled photons and some degree of nonlocality. Extreme non-degenerate photons are born together in optical parametric oscillator. They can contain a large fraction of entangled photons in contrast to degenerate photons [16]. Experiments with the Hong-Ou-Mandel effect would be interesting here [17]. The absorption of entangled photons in a semiconductor is a reversed (or partially reversed) process with a very large differential cross-section [18, 19].

The purpose of this note is to draw the attention of physicists to the results of colleagues and the need for an experimental study of nonlocality in quantum physics. This non-locality is obviously present in experiments with ETPA, but this is easier to study in experiments with light splitters [16, 20].

- B. Dayan, A. Pe'er, A. A. Friesem, and Y. Silberberg, "Two Photon Absorption and Coherent Control with Broadband Down-Converted Light", Phys. Rev. Lett. 93, 023005 (2004).
- D. I. Lee and T. Goodson III, "Entangled Photon Absorption in an Organic Porphyrin Dendrimer", J. Phys. Chem. B, 110, 25582 (2006).
- J. P. Villabona-Monsalve, O. Calderón-Losada, M. Nuñez Portela, and A. Valencia, "Entangled Two Photon Absorption Cross Section on the 808 nm Region for the Common Dyes Zinc Tetraphenylporphyrin and Rhodamine B", J. Phys. Chem. A, **121**, 7869 (2017).
- D. Tabakaev, M. Montagnese, G. Haack, L. Bonacina, J.-P. Wolf, H. Zbinden, R. T. Thew, "Energy-Time Entangled Two-Photon Molecular Absorption ", e-print, arXiv:1910.07346.
- O. Varnavski and T. Goodson III, "Two-Photon Fluorescence Microscopy at Extremely Low Excitation Intensity: The Power of Quantum Correlations", J. Am. Chem. Soc., 142, 12966 (2020).
- K. M. Parzuchowski, A. Mikhaylov, M. D. Mazurek, R. N. Wilson, D. J. Lum, T. Gerrits, C. H. Camp Jr., M. J. Stevens, and R. Jimenez, "Setting bounds on two-photon absorption cross-sections in common fluorophores with entangled photon pair excitation", e-print, arXiv:2008.02664.
- T. Landes, M. Allgaier, S. Merkouche, B. J. Smith, A. H. Marcus, and M. G. Raymer, "Experimental feasibility of molecular two-photon absorption with isolated timefrequency-entangled photon pairs", e-print, arXiv:2012.06736.
- S. Corona-Aquino, O. Calderón-Losada, M. Y. Li-Gómez, H. Cruz-Ramirez, V. Alvarez-Venicio, M. P. Carreón-Castro, R. J. León-Montiel, and A. B. U'Ren, "Experimental study on the effects of photon-pair temporal correlations in entangled two-photon absorption", e-print, arXiv:2101.10987.
- 9. V. A. Kuz'menko, "Comment on entangled two-photon absorption in molecules", eprint, viXra:2103.0014.
- D. A. Fishman, C. M. Cirloganu, S. Webster, L. A. Padilha, M. Monroe, D. J. Hagan, and E. W. Van Stryland, "Sensitive mid-infrared detection in wide-bandgap semiconductors using extreme non-degenerate two-photon absorption", Nat. Photon. 5, 561 (2011).
- Y. Zhang, C. Husko, S. Lefrancois, I. H. Rey, T. F. Krauss, J. Schröder, and B. J. Eggleton, "Non-degenerate twophoton absorption in silicon waveguides: Analytical and experimental study", Opt. Express 23, 17101 (2015).

- J. Fang, Y. Wang, M. Yan, E. Wu, K. Huang, and H. Zeng, "Highly Sensitive Detection of Infrared Photons by Nondegenerate Two-Photon Absorption Under Midinfrared Pumping", Phys. Rev. Applied 14, 064035 (2020).
- N. Cox, J. Wei, H. Pattanaik, T. Tabbakh, S. P. Gorza, D. Hagan, and E. W. Van Stryland, "Nondegenerate Two-Photon Absorption in GaAs/AlGaAs Multiple Quantum Well Waveguides", e-print, arXiv:1912.01685.
- 14. D. Knez, A. M. Hanninen, R. C. Prince, E. O. Potma, and D. A. Fishman, "Infrared chemical imaging through nondegenerate two-photon absorption in silicon-based cameras", Light Sci. Appl. 9, 125 (2020).
- 15. E. O. Potma, D. Knez, Y. Chen, A. Durkin, A. Fast, M. Balu, B. Norton-Baker, T. Baldacchini, R. Martin, D. A. Fishman, "Rapid chemically selective 3D imaging in the mid-infrared with a Si-based camera", e-print, arXiv:2103.01159.
- V. A. Kuz'menko, "On the physical nature of Hanbury-Brown-Twiss and Hong-Ou-Mandel effects", e-print, viXra:2012.0188
- 17. H. Chen, T. Peng, and Y. Shih, "100% correlation of chaotic thermal light", Phys. Rev., A 88, 023808 (2013).
- 18. B. Dayan, A. Pe'er, A. A. Friesem, and Y. Silberberg, "Coherent control with broadband squeezed vacuum", e-print, arXiv:quant-ph/302038.
- 19. V. A. Kuz'menko, "Time reversal noninvariance in quantum physics", e-print, viXra:2004.0160.
- 20. V. A. Kuz'menko, "On the experimental study of nonlocality in quantum physics", eprint, viXra:1902.0331.