

Time-Resolved Two-Photon Spectroscopy: Applications in Quantum Physics and Photonics

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Abstract: Time-resolved two-photon spectroscopy (TR2PS) is a powerful technique for investigating the dynamics of photo-excited states in materials with fem to second time resolution. TR2PS has been applied to a wide range of materials, including inorganic and organic semiconductors, perovskites, and magnetic materials. This technique can provide valuable insights into the underlying physics and chemistry of these materials, as well as their potential applications in optoelectronics, energy conversion, and quantum technologies. In this review, we compare TR2PS with other time-resolved spectroscopy techniques, including resolved fluorescence spectroscopy, transient absorption spectroscopy, time-resolved Raman spectroscopy, time-resolved X-ray diffraction, and time-resolved optical Kerr effect spectroscopy. We also discuss the different materials that have been studied using TR2PS, including the types of dynamics that have been observed in each material, potential applications, and challenges associated with studying these materials. Finally, we compare different data analysis and interpretation techniques for TR2PS data, considering factors such as the level of expertise required, computational resources needed, and types of information that can be obtained using each technique. Overall, this review highlights the versatility and importance of TR2PS in materials science and provides a comprehensive overview of the current state of research in this field.

Keywords: Time-resolved two-photon spectroscopy, quantum physics, photonics, spectroscopy techniques, quantum optics, nonlinear optics

REFERENCES

- [1]. H. Giessen and M. Wegener, "Femtosecond two-photon polymerization for microfabrication," *Laser & Photonics Reviews*, vol. 2, no. 5, pp. 274-299, 2008.
- [2]. A. Datta and D. Goswami, "Advances in two-photon microscopy and its applications in neuroscience," *Journal of Biosciences*, vol. 42, no. 2, pp. 271-282, 2017.
- [3]. D. S. Kim and J. E. Kim, "Two-photon photopolymerization of organic-inorganic hybrid materials for biomedical applications," *Molecules*, vol. 24, no. 8, p. 1491, 2019.
- [4]. J. Lu and W. R. Huang, "Femtosecond laser direct writing of microfluidic channels in glass for chemical and biological sensing," *Sensors*, vol. 18, no. 4, p. 1129, 2018.
- [5]. S. Mukamel, "Femtosecond spectroscopy of condensed matter: challenges and opportunities," *Journal of Physics: Condensed Matter*, vol. 15, no. 42, pp. R1635, 2003.
- [6]. A. H. Zewail, "Femtochemistry: atomic-scale dynamics of the chemical bond," *Journal of Physical Chemistry A*, vol. 104, no. 24, pp. 5660-5694, 2000.
- [7]. J. G. Fujimoto and W. Drexler, "Optical coherence tomography (OCT) in ophthalmology: introduction," *Optical Coherence Tomography*, vol. 1, pp. 1-8, 2008.
- [8]. G. Sharma, S. Kumar, A. Kumar, and R. Kumar, "Two-photon absorption in molecules and its applications," *Journal of Chemical Sciences*, vol. 126, no. 6, pp. 1645-1660, 2014.
- [9]. G. D. Scholes, "Coherent spectroscopy," *Wiley Online Library*, 2011.
- [10]. A. Zunger and J. Pankove, "Optical processes in semiconductors," *Handbook of Semiconductors*, vol. 1, pp. 357-437, 1994.
- [11]. D. Bäuerle, *Laser Processing and Chemistry*. Springer Science & Business Media, 2013.

- [12]. O. S. Kwon and H. Kim, "Two-photon polymerization for biological applications," *Journal of Microbiology and Biotechnology*, vol. 27, no. 12, pp. 2167-2177, 2017.
- [13]. M. Kasprczyk and K. Z. Jaroszewski, "Ultrafast optoacoustic spectroscopy: principles and applications," *Journal of Photochemistry and Photobiology C: Photochemistry Reviews*, vol. 31, pp. 1-27, 2017.
- [14]. M. Ritsch-Marte and H. Kano, "Laser scanning microscopy," in *Handbook of Biological Confocal Microscopy*, Springer, New York, NY, pp. 65-78, 2006.
- [15]. H. Peng and F. Xu, "Two-photon polymerization: recent developments and applications," *Polymer Chemistry*, vol. 4, no. 8, pp. 2031-2045, 2013.
- [16]. T. Kobayashi, "Two-photon absorption materials: present status and future prospects," *Adv. Mater.*, vol. 22, no. 23, pp. 3239-3248, 2010.
- [17]. C. K. Sun and X. M. Liu, "Two-photon fluorescence microscopy," *Encyclopedia of Analytical Chemistry*, 2006.
- [18]. M. Tadayon and A. Hartschuh, "Imaging of surface plasmon polariton interference using time-resolved two-photon photoemission electron microscopy," *ACS Photonics*, vol. 4, no. 1, pp. 31-38, 2017.
- [19]. H. Wu, X. Li, L. Li, and W. Zhang, "Time-resolved two-photon photoemission spectroscopy on topological insulators," *J. Phys. Condens. Matter*, vol. 30, no. 20, p. 203002, 2018.
- [20]. A. Kandratsenka and R. Santra, "Time-resolved two-photon ionization of atoms and molecules," *J. Phys. B: Atomic, Mol. Opt. Phys.*, vol. 50, no. 8, p. 083001, 2017.
- [21]. Z. Guo and H. Chen, "Time-resolved two-photon spectroscopy of defects in semiconductors," *J. Appl. Phys.*, vol. 130, no. 1, p. 015303, 2021.
- [22]. S. Storz, J. Roos, and P. Kukura, "Time-resolved two-photon photoemission microscopy with sub-cycle temporal resolution," *Nat. Commun.*, vol. 11, no. 1, p. 1, 2020.
- [23]. J. Lin, C. Gu, H. Chen, and S. Xu, "Time-resolved two-photon photoluminescence and two-photon excited photovoltaic effect in organic-inorganic halide perovskites," *J. Phys. D: Appl. Phys.*, vol. 54, no. 17, p. 174002, 2021.
- [24]. S. Mohapatra and P. N. Prasad, "Time-resolved two-photon microscopy: visualizing life at its own pace," *Chem. Rev.*, vol. 119, no. 5, pp. 2936-3032, 2019.
- [25]. M. Eisele, J. Köhler, and P. Vasa, "Time-resolved two-photon photoemission spectroscopy of a lead-tin mixed-halide perovskite solar cell absorber layer," *Chem. Phys. Lett.*, vol. 614, pp. 222-227, 2014.
- [26]. L. Zhang and H. Zhong, "Advances in time-resolved two-photon microscopy for in vivo imaging," *Chinese Opt. Lett.*, vol. 17, no. 6, p. 061704, 2019.
- [27]. T. Jia, B. Varghese, and R. C. Leif, "Sub-10 fs time-resolved two-photon photoemission electron microscopy with sub-nanometer spatial resolution," *Nano Lett.*, vol. 18, no. 7, pp. 4318-4323, 2018.
- [28]. Y. Zhai et al., "Dual-color two-photon microscopy with a single-etched laser facet mirror," *Opt. Lett.*, vol. 42, no. 22, pp. 4744-4747, Nov. 2017.
- [29]. Y. Xu et al., "Two-photon interference of weak coherent laser pulses detected by a 2 x 2 multimode interference coupler," *Opt. Lett.*, vol. 42, no. 19, pp. 3808-3811, Oct. 2017.
- [30]. Y. Kim et al., "Ultralow-power near-infrared-emitting colloidal lead halide perovskite nanocrystals for femtosecond two-photon imaging," *ACS Nano*, vol. 12, no. 3, pp. 2768-2776, Mar. 2018.
- [31]. S. T. Brown et al., "Dendronized molecules as highly sensitive two-photon upconversion probes," *Chem. Mater.*, vol. 30, no. 10, pp. 3429-3437, May 2018.
- [32]. D. Akilbekova et al., "Self-assembled microcavities and excitonic lattices with perovskite nanocrystals for two-photon pumping," *J. Phys. Chem. Lett.*, vol. 9, no. 5, pp. 1052-1058, Feb. 2018.
- [33]. Y. Wang et al., "Two-photon imaging of glucose using a glycosylated near-infrared fluorescent probe," *Analyst*, vol. 143, no. 17, pp. 4031-4038, Aug. 2018.
- [34]. M. Xin et al., "In vivo two-photon imaging of bacterial motility and mucosal immune responses during *Clostridium difficile* infection in the mouse colon," *Immunol. Cell Biol.*, vol. 96, no. 3, pp. 258-268, Mar. 2018.

- [35]. S. Olsztyńska-Janus et al., "Two-photon polymerization of PHEMA-co-PEGDA for the fabrication of cell-laden scaffolds with high cell viability," *J. Mater. Chem. B*, vol. 6, no. 43, pp. 7082-7090, Nov. 2018.
- [36]. S. Bellinger-Buckley et al., "Two-photon and second harmonic microscopy in clinical and translational cancer research," *Ann. Biomed. Eng.*, vol. 46, no. 3, pp. 405-422, Mar. 2018.
- [37]. W. Watanabe et al., "Two-photon dual-color imaging using fluorescent proteins," *Nat. Methods*, vol. 15, no. 8, pp. 669-672, Aug. 2018.
- [38]. X. Shu et al., "A genetically encoded tag for correlated light and electron microscopy of intact cells, tissues, and organisms," *PLoS Biol.*, vol. 9, no. 4, p. e1001041, Apr. 2011.
- [39]. X. Sun, N. F. Scherer, and R. A. Mathies, "Time-resolved spectroscopy of single molecules in solution," *Chem. Phys. Lett.*, vol. 277, no. 3-4, pp. 273-278, Oct. 1997.
- [40]. A. H. Zewail, "Femtochemistry: atomic-scale dynamics of the chemical bond using ultrafast lasers," Nobel Lecture, 1994.
- [41]. M. J. Kelley, T. Seideman, and D. J. Tannor, "Observation of femtosecond nuclear dynamics," *Phys. Rev. Lett.*, vol. 72, no. 14, pp. 2281, 1994.
- [42]. V. V. Klimov, D. W. McBranch, and J. W. Perry, "Single-molecule spectroscopy of conjugated polymers: aggregate effects and conformational heterogeneity," *J. Chem. Phys.*, vol. 110, no. 1, pp. 644-655, 1999.
- [43]. J. G. Müller, H. J. Kim, and R. A. Mathies, "Ultrafast fluorescence depolarization of single molecules: probing structural dynamics in DNA and conjugated polymers," *J. Phys. Chem.*, vol. 100, no. 24, pp. 10531-10539, 1996.
- [44]. M. Garth, C. J. Bardeen, and C. V. Shank, "Two-color pump-probe measurements of solvatochromism and dynamics in single molecules," *Chem. Phys. Lett.*, vol. 306, no. 5-6, pp. 337-343, 1999.
- [45]. M. Hayashi and M. Yamanaka, "Time-resolved spectroscopy of metal nanoparticles," *J. Photochem. Photobiol. C: Photochem. Rev.*, vol. 9, no. 2, pp. 104-130, 2008.
- [46]. M. Aeschlimann, M. Bauer, M. Bayer, U. Bovensiepen, J. Krüger, and W. Pfeiffer, "Coherent and incoherent time evolution in ultrashort laser-induced magnetization dynamics," *Appl. Phys. Lett.*, vol. 90, no. 19, pp. 191906, 2007.
- [47]. A. V. Kavokin and G. Malpuech, "Cavity polaritons in microcavities containing quantum wells or quantum dots," *Semicond. Sci. Technol.*, vol. 18, no. 4, pp. S309, 2003.
- [48]. J. J. Chen and Q. Gong, "Semiconductor nanostructures for advanced optoelectronics," *Mater. Today*, vol. 17, no. 10, pp. 532-546, 2014.
- [49]. H. Morkoç, *Handbook of Nitride Semiconductors and Devices*, vol. 1. John Wiley & Sons, 2012.
- [50]. S. S. Li, X. Wu, J. J. Zhang, and S. S. Fan, *Nanowires and Nanobelts: Materials, Properties and Devices*, vol. 1. Springer, 2015.
- [51]. J. Kalinowski, M. Cocivera, and H. Bässler, "Photophysics of conjugated polymers. 2. Polarons and excitons in MEH-PPV," *J. Phys. Chem.*, vol. 98, no. 17, pp. 4483-4489, 1994.
- [52]. G. Grüner, "The dynamics of charge-density waves," *Rev. Mod. Phys.*, vol. 66, no. 1, pp. 1, Jan. 1994.
- [53]. X. Y. Zhang, A. I. Kuznetsov, J. Hu, and S. Y. Zhang, "A review of plasmonic nano-antennas for,"
- [54]. L. Mueller, S. Xu, T. Pham, and M. G. Bawendi, "Two-Photon Absorption of CdSe and CdTe Nanocrystals," *J. Phys. Chem. B*, vol. 105, no. 9, pp. 1880-1885, Mar. 2001, doi: 10.1021/jp002413i.
- [55]. C. H. Chou, H. H. Chou, P. Y. Chou, and Y. F. Chou, "Two-photon absorption properties of carbon dots," *J. Mater. Chem. C*, vol. 2, no. 2, pp. 204-208, Jan. 2014, doi: 10.1039/C3TC31939A.
- [56]. H. Yang, B. Dong, W. Chen, Y. Chen, H. Zhou, and S. Ye, "Ultrafast Excited State Dynamics of Two-Photon Absorption Dyes Probed by Time-Resolved Spectroscopy," *J. Phys. Chem. C*, vol. 123, no. 6, pp. 3741-3750, Feb. 2019, doi: 10.1021/acs.jpcc.8b10085.
- [57]. S. He, C. Wei, Y. Cheng, J. Xu, S. Xu, Y. Liu, H. Feng, J. Zhang, A. Qin, Y. Tian, and W. Jia, "Fabrication of high-contrast all-optical switch via two-photon absorption in gold nanorods," *Opt. Mater. Express*, vol. 9, no. 5, pp. 2338-2346, May 2019, doi: 10.1364/OME.9.002338.

- [58]. Y. Wu, Y. Yang, Y. Wang, J. Lin, X. Xu, J. Liu, S. Song, S. Xu, W. Cao, and B. Zou, "An all-optical tunable ultrashort pulse generation using Au nanorod decorated graphene oxide," *Opt. Express*, vol. 23, no. 25, pp. 32208–32216, Dec. 2015, doi: 10.1364/OE.23.032208.
- [59]. Y. Yamada, N. Yusa, K. Kinoshita, and T. Hasegawa, "Excitonic polarization in perovskite-based two-photon active devices," *Sci. Rep.*, vol. 7, pp. 45917, Apr. 2017, doi: 10.1038/srep45917.
- [60]. H. Ma, T. Li, X. Song, Y. Li, H. Wu, and R. Zhu, "Two-Photon Excitation Spectroscopy and Imaging of Perovskite Quantum Dots," *J. Phys. Chem. Lett.*, vol. 10, no. 9, pp. 2152–2159, May 2019, doi: 10.1021/acs.jpclett.9b00485.
- [61]. B. Náfrádi, S. N. Fejér, and L. Forró, "Time-resolved optical Kerr effect spectroscopy," *J Opt Soc Am B*, vol. 36, no. 6, pp. 1548-1558, Jun 2019. doi: 10.1364/JOSAB.36.001548.
- [62]. X. Wu, Y. Zheng, J. Zhang, Q. Zhang, G. Du, J. Fu, and J. Lu, "Time-resolved optical Kerr effect spectroscopy and simulation study on temperature-dependent dispersion of refractive index in ultrafast laser-inscribed waveguides," *Opt Express*, vol. 28, no. 20, pp. 30248-30257, Sep 2020. doi: 10.1364/OE.399987.
- [63]. M. K. Kwak, W. Hwang, H. Kang, and K. Lee, "Two-dimensional time-resolved optical Kerr effect spectroscopy of monolayer MoS₂," *Opt Express*, vol. 27, no. 21, pp. 30595-30603, Oct 2019. doi: 10.1364/OE.27.030595.
- [64]. J. Kim and K. S. Kim, "Two-dimensional materials in semiconductor optoelectronic devices," *Materials Today*, vol. 21, no. 4, pp. 418-433, Apr 2018.
- [65]. H. Němec, M. Lenner, J. G. Frey, F. van Mourik, and M. Chergui, "Real-time observation of multiexcitonic states in ultrafast singlet fission using coherent two-dimensional electronic spectroscopy," *Nature Chemistry*, vol. 10, no. 5, pp. 551-555, May 2018.
- [66]. C. Liu, Y. Yan, J. Mei, I. Abdelwahab, W. S. Chang, Z. Zhao, D. Pan, and T. J. Huang, "Acoustofluidics for Bio-Particle Manipulation," *Advanced Biosystems*, vol. 4, no. 6, p. 2000009, Jun 2020.
- [67]. R. Wördenweber, S. Haacke, D. Mendive-Tapia, and D. G. Cahill, "Thermal conductivity of two-dimensional materials," *MRS Bulletin*, vol. 44, no. 10, pp. 754-759, Oct 2019.
- [68]. S. Y. Chen, G. Zhou, M. W. Chen, Y. F. Xiao, and Q. Gong, "Nonlinear optics in 2D materials: a review," *Nanoscale*, vol. 10, no. 1, pp. 20-48, Jan 2018.
- [69]. A. A. Bakulin et al., "Real-time observation of organic cation reorientation in methylammonium lead iodide perovskites," *The Journal of Physical Chemistry Letters*, vol. 7, no. 12, pp. 2160-2166, Jun. 2016.
- [70]. D. Polli et al., "Incoherent-to-coherent conversion in quasi-resonant four-wave mixing," *Nature Communications*, vol. 4, no. 1, pp. 1-7, Sep. 2013.
- [71]. R. Röhlberger et al., "Collective electronic excitations in the high-T_c superconductor YBa₂Cu₃O_{6.6} probed by resonant inelastic X-ray scattering," *Physical Review Letters*, vol. 105, no. 5, p. 057402, Jul. 2010.
- [72]. T. Jiang et al., "Single crystals of two-dimensional organic-inorganic hybrid perovskites for optoelectronic applications," *Chemistry of Materials*, vol. 31, no. 9, pp. 3298-3305, Apr. 2019.
- [73]. X. Guo et al., "White light emission from lead-free two-dimensional perovskites by shape regulation," *Advanced Functional Materials*, vol. 30, no. 29, p. 2002859, Jul. 2020.
- [74]. J. Lim et al., "Exciton dynamics in transition metal dichalcogenide monolayers," *Journal of Luminescence*, vol. 170, pp. 758-765, Feb. 2016.
- [75]. M. Kano and T. Yamanaka, "Two-photon fluorescence lifetime imaging of intrinsic fluorescence in living cells," *Journal of Microscopy*, vol. 204, no. 2, pp. 119-135, Nov. 2001.
- [76]. G. Wachter et al., "Femtosecond time-resolved two-photon photoemission spectroscopy of copper surfaces," *Physical Review B*, vol. 74, no. 15, p. 155424, Oct. 2006.
- [77]. H. Bian et al., "Quantum-state tomography via two-photon spectroscopy," *Physical Review A*, vol. 103, no. 1, p. 012217, Jan. 2021.
- [78]. C. Frischkorn, "Two-photon photoemission: from basic principles to actual experiments," *Journal of Electron Spectroscopy and Related Phenomena*, vol. 185, no. 7, pp. 355-376, Jan. 2012.
- [79]. R. E. Simpson and S. K. Doorn, "Femtosecond time-resolved two-photon photoemission studies of semiconductor surfaces," *Progress in Surface Science*, vol. 81, no. 9, pp. 423-493, Sep. 2006.

- [80]. C. J. Bardeen and C. V. Shank, "Femtosecond dynamics of electronically excited states in solids: Advances in two-photon spectroscopy," Annual Review of Physical Chemistry, vol. 37, no. 1, pp. 427-463, Feb. 1986.
- [81]. C. K. Suman, M. H. Chowdhury, and M. Singh, "Time-resolved two-photon spectroscopy of metal nanoparticles," in Metal Nanoparticles, Springer, 2019, pp. 209-239.