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Theory of time-resolved optical spectroscopy with quantum entangled photons

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Abstract

Quantum light exhibits unique photon statistics and correlations absent in classical light. Harnessing these quantum properties allows selective excitation and precise measurements that cannot be performed in spectroscopic measurements using classical light such as lasers. With advances in quantum optical technology, there has been increased interest in the use of quantum light in nonlinear spectroscopic measurements of molecular systems. In these respects, applying entangled photons to time-resolved spectroscopy can open new avenues for unambiguously extracting information on dynamical processes in complex molecular and material systems.

We propose transmission measurement of frequency-entangled broadband photon pairs generated via parametric down-conversion with a monochromatic laser [1,2]. It is observed that state-to-state dynamics in the system under study are temporally resolved by adjusting the path difference between the entangled twin beams when the entanglement time is sufficiently short. The non-classical photon correlation enables time-resolved spectroscopy with monochromatic pumping.

Furthermore, we consider a quantum spectroscopy measurement method extending the above time-resolved quantum spectroscopy to a two-photon coincidence type [3]. We demonstrate that the photon number correlation enables the selective elimination of specific signal pathways in nonlinear spectra, which cannot be realized with classical coherent light. We anticipate that the proposed spectroscopy will help simplify the spectral interpretation of complex molecular and material systems comprising multiple molecules. Finally, we discuss how time-resolved quantum spectroscopy can be

achieved using current photon detection technology [4].

[1] A. Ishizaki, *J. Chem. Phys*. **153**, 051102 (2020).
[2] Y. Fujihashi, K. Miwa, M. Higashi, A. Ishizaki, *J. Chem. Phys*. **159**, 114201 (2023).
[3] Y. Fujihashi, A. Ishizaki, and R. Shimizu, *J. Chem. Phys*. **160**, 104201 (2024).
[4] Y. Fujihashi, O. Iso, A. Ishizaki, and R. Shimizu, *in preparation*.