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Higher-order correlations uncover hidden information in the fluorescence fluctuation analysis of fast molecular kinetics FARSHAD ABDOLLAH-NIA, MARTIN GELFAND, Department of Physics, Colorado State University, ALAN VAN ORDEN, Department of Chemistry, Colorado State University — The statistical analysis of photons collected from fluorescent molecules as they diffuse and react in solution can provide information about concentrations and reaction rates. Two common techniques, based on correlation (FCS) and histogram (PCH) analysis of photon statistics, are unable to provide a complete picture of fast molecular reactions. Higher-order correlations (HOFCS) can, in principle, overcome such limitations. Historically, attempts to use HOFCS have been hindered by modeling complications in theory, and by shot noise and detector artifacts in experiment. Melnykov and Hall (2009) solved the modeling problem through a cumulant-based formulation of HOFCS [1]. More recently, we have introduced techniques to evaluate artifact-free higher-order correlation functions with improved time resolution, overcoming the experimental limitations [2]. This has enabled us to apply HOFCS to fast reversible reactions for the first time [3].

[1] A. V. Melnykov and K. B. Hall, The Journal of Physical Chemistry B 113, 15629 (2009)
[2] F. Abdollah-Nia, M. P. Gelfand, and A. Van Orden, The Journal of Physical Chemistry B 121, 2373 (2017)
[3] F. Abdollah-Nia, M. P. Gelfand, and A. Van Orden, The Journal of Physical Chemistry B 121, 2388 (2017)

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