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Theory for Frequency-Resolved Photon Emission Statistics in Single-Molecule Fluorescence Spectroscopy GOLAN BEL, Center for Nonlinear Studies and CCS-3, Los Alamos National Laboratory, FRANK BROWN, Department of Chemistry and Biochemistry and Department of Physics, University of California, Santa Barbara — We derive the moment generating function for photon emission from a single molecule driven by laser excitation. The frequencies of the fluoresced photons are explicitly considered. Calculations were performed for the case of a two level dye molecule, showing that measured photon statistics will display a strong and nonintuitive dependence of detector bandwidth. Moreover it is demonstrated that the anti-bunching phenomenon associated with negative values of Mandel's Q parameter, results from correlations between photons with well separated frequencies.

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