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Electrostatic Steering of Functional Dynamics in GFP J. TIMOTHY SAGE, Northeastern Univ., GEORGI Y. GEORGIEV, Assumption College, JASPER J. VAN THOR, University of Oxford — Distinctive photodynamic properties of the green fluorescent protein (GFP) from the jellyfish *A. victoria* result from charge transfer processes involving the autocatalytically generated chromophore. We investigate structural changes in response to chromophore photoionization at cryogenic temperatures, using both X-ray crystallography and polarized infrared measurements on oriented single crystals. These measurements identify conformational changes of Gln 69, Cys 70, and an associated H-bonded cluster of internal water molecules in the chromophore environment. These structural changes take place at 100 K, far below the "dynamical transition" traditionally regarded as enabling functional protein motions. This contrasts with the prevailing view that the rigid interior of the GFP β-barrel sterically inhibits nonradiative processes. Instead, we propose that rapid rearrangements of the chromophore environment enhance the fluorescence quantum yield by stabilizing the abruptly altered charge distribution in the radiative state. We suggest that the conformational response to charge transfer influences two fundamental and useful spectroscopic properties of GFP—the large frequency separation between excitation and emission and the efficient fluorescence.

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