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Femtosecond Nonlinear Optical Studies of Radiationless Decay in Carotenoids and in the Peridinin–Chlorophyll *a* Protein[1] SOUMEN GHOSH, MICHAEL BISHOP, JENNY JO MUELLER, NOLAN SHEPHERD, WARREN BECK, Michigan State Univ, HARRY FRANK, University of Connecticut — Femtosecond transient-grating spectroscopy with optical heterodyne detection was employed to observe the time evolution of the absorption and dispersion components of the third-order nonlinear optical signal following resonant excitation of the S_2 ($1B_u^+$) states of βcarotene in benzonitrile and peridinin in methanol. The absorption and dispersion components exhibit distinct time profiles owing to the population of dark intermediate states. An initial intermediate is populated on an ultrashort (<30 fs) time scale in both carotenoids owing to the onset of torsional distortions on the S_2-state potential surface. The time-resolved transient-grating spectra obtained for peridinin in the peridinin–chlorophyll *a* protein from *Amphidinium carterae* indicate that the intermediate is formed even more rapidly than in solution. This finding suggests that the twisted conformation of the peridinin chromophore is controlled in the binding site so as to optimize energy transfer to chlorophyll *a* by enhancing the formation of an intramolecular charge-transfer character.

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