Department of Chemistry and Biochemistry
Physical Chemistry Seminar Series

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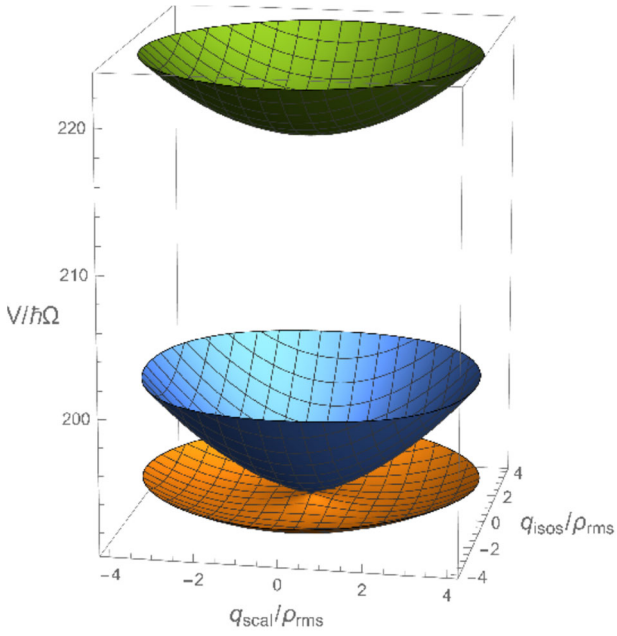
Tykeson 140

Interference, coherence, and Berry's phase development in an excitation-transfer trimer.

Abstract: I will discuss several interesting features in the dynamics of an equilaterally shaped electronic excitation-transfer (EET) trimer with distance-dependent intermonomer couplings. This trimer is a model for circular photosynthetic light-harvesting complexes. Of specific interest are the effects of multi-path interference in EET to one monomer, starting from symmetric and antisymmetric superpositions of single-monomer excitations on the other two. In its equilateral triangular configuration, two of the three singly electronically excited states are degenerate. This degeneracy is broken by the Jahn-Teller-active framework distortions.



Calculations illustrate closed, approximately circular pseudo-rotational wave-packet dynamics on both the lower and the upper adiabatic potential energy surfaces of the degenerate manifold, which lead to the acquisition after one cycle of physically meaningful geometric (Berry) phases of 180 degrees. A vivid indirect manifestation of Berry-phase development—recently observed in remarkable quantum-simulations on trapped atomic ions—is seen in the evolution of the probability density on the lower Jahn-Teller potential of a wave packet comprising a superposition of clockwise and counterclockwise circular motion. I'll briefly explain prospective strategies for initiating and monitoring these various dynamical processes using pre-resonant impulsive Raman excitation, short-pulse absorption, and multidimensional wave-packet interferometry, and suggest a specific trimeric target in the form of a tripodal tetracene-based analog of triptycene.