New Insights into Hydrogen Tunneling in Soybean Lipoygenase-1 using Quantum Wavepacket 

Abstract

We study the hydrogen tunneling problem in the two-state lipoygenase-1 using quantum transport dynamics under the influence of a classical active site (AS). The AS includes proton donor and acceptor groups, the tunneling potential energy surfaces and the tunneling probability are found. Our model uses an optimal geometry to describe the quantum wavepacket evolution during the tunneling process. The changing AS geometry causes a sequence of quantum events that drive a donor localized wavepacket onto an extended state in the product side. This process has consequences with quantum measurement theory.

Introduction

Many examples where hydrogen tunneling might play a critical role in enzyme-catalyzed biological reactions can be found in the literature. Evidence supporting this hypothesis is shown in the current study. The potential energy surface of the enzyme-catalyzed reactions is complex, and the individual-state quantum dynamics and the tunneling probability are found. Our model uses an optimal geometry to describe the quantum wavepacket evolution during the tunneling process. The changing AS geometry causes a sequence of quantum events that drive a donor localized wavepacket onto an extended state in the product side. This process has consequences with quantum measurement theory.

Active Site and Wavepacket Dynamics

In our model, we consider the hydrogen tunneling in the enzyme-catalyzed reactions and model the anions and liquids [see Figure 1]. We also used ketohydrazone to model the substrate, linoleic acid [see Figure 2].

Figure 1. The left figure shows the active site encapsulated in the NADH enzyme and the right figure displays the molecular system. In order to calculate a reaction pathway, we:

- performed a relaxed run along the reaction coordinate defined as (RCH - ROH)/RCO, where C is the donor carbon, O is the acceptor oxygen, and H is the shared hydrogen.
- calculated the tunneling hydrogen potential energies on each reaction coordinate at each step along the reaction coordinate.
- the potential energy and optimization at each step were calculated with BLYP/LANL2DZ. Each nucleus is a smoothly variable with the donor state minimizing increasing and the acceptor state minimizing decreasing along the energy profile shown in Figure 1. This technique approximates the classical behavior of the active site.

The time-evolution of the quantum wavepacket is obtained through Trotter symmetric factorization of the quantum propagator. The accumulated KIE (KIE) is approximated using the wavepacket transmission coefficient (see Figure 2). The time-evolution of the quantum wavepacket is obtained through Trotter symmetric factorization of the quantum propagator.

Conclusions

In summary, we have:

- provided correlation between for S0 and generated a hydrogen potential energy at points along the profile
- propagated a quantum tunneling with a time-dependent Hamiltonian (time-dependent potential)
- measured eigenvalues at positions in particular for quantum levels
- explored the experimental KIE for S0 without any further analysis using wavepacket propagation
- connected the tunneling process to measurement theory where the active site geometry as a measuring device that drive the tunneling particle from a donor to acceptor state
- shown that $|\langle M \rangle|/|\langle H \rangle|$ increases along with the wavepacket transmission coefficient across the reaction profile, suggesting the active site dynamics increase the tunneling probability during the reaction

The quantity of the inverse temperature. Since this quantity grows monotonically with $|\langle M \rangle|$ and $|\langle H \rangle|$, it is apparent that the projection of the quantum states on the tunneling is also important in absolute value.

References