The Journal of Physical Chemistry C > Vol 129/Issue 31 > Article

Subscribed

PERSPECTIVE | June 17, 2025

Density Functional Tight-Binding Enables Tractable Studies of Quantum Plasmonics

Nikhil S. Chellam, Subhajyoti Chaudhuri, Abhisek Ghosal, Sajal Kumar Giri*, and George C. Schatz*

Open PDF

Supporting Information (1)

Abstract

Routine investigations of plasmonic phenomena at the quantum level present a formidable computational challenge due to the large system sizes and ultrafast time scales involved. This Feature Article highlights the use of density functional tight-binding (DFTB), particularly its real-time time-dependent formulation (RT-TDDFTB), as a tractable approach to study plasmonic nanostructures from a quantum mechanical purview. We begin by outlining the theoretical framework and limitations of DFTB, emphasizing its efficiency in modeling systems with thousands of atoms over picosecond time scales. Applications of RT-TDDFTB are then explored in the context of optical absorption, nonlinear harmonic generation, and plasmon-mediated photocatalysis. We demonstrate how DFTB can reconcile classical and quantum descriptions of plasmonic behavior, capturing key phenomena such as size-dependent plasmon shifts and plasmon coupling in nanoparticle assemblies. Finally, we showcase DFTB's ability to model hot carrier generation and reaction dynamics in plasmon-driven H_2 dissociation, underscoring its potential to model photocatalytic processes. Collectively, these studies establish DFTB as a powerful, yet computationally efficient tool to probe the emergent physics of materials at the limits of space and time.