

Understanding the structure-property relationship of ACC-Dimers via high-throughput density functional theory and machine learning

Giacomo G. Nagaro,^a Luke Nambi Mohanam,^b Peter M. Mastracco,^c Sangram Prusty,^a
Younghoon Oh,^a Qiang Cui,^a Stacy Copp,^{cde} Sahar Sharifzadeh^{bfa}

Department of Chemistry, Boston University^a

Department of Electrical and Computer Engineering, Boston University^b

Department of Materials Science and Engineering, University of California, Irvine^c

Department of Chemical and Biomolecular Engineering, University of California, Irvine^d

Department of Physics and Astronomy, University of California, Irvine^e

Department of Physics, Boston University^f

We present an all-atom molecular dynamics (MD) and density functional theory (DFT) study of the explicitly solvated antiparallel coiled-coiled dimer (ACC), a self-assembled helical peptide bundle formed by two 29-residue α -helix peptide chains. ACC biofilms have been shown experimentally to efficiently conduct electricity and are therefore promising as components in synthetic bioelectronic materials. Classical MD under ambient conditions captures the thermal nuclear vibrations of this system. DFT calculations of 1000 MD snapshots of the ACC-Dimer with an explicit water solvation shell show near-degenerate frontier Kohn-Sham orbitals, which we expect to be the main driver of conductivity, that fluctuate with nuclear vibrations. Such a finding is consistent with a vibration-induced conductivity model. To understand this observation, we utilize feature analysis and machine learning algorithms to understand the relationship between nuclear geometry and the number of near-degenerate frontier orbitals. These results demonstrate the importance of a full atomistic picture for studying the electronic properties of supramolecular protein complexes.