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Title:Long-time correlations and hydrophobe-modified hydrogen-bonding dynamics in hydrophobic hydration
Authors:Titantah, John Tatini (1); Karttunen, Mikko (2)
Author affiliation:(1) Department of Applied Mathematics, University of Western Ontario, London, ON N6A 5B7, Canada; (2) Department of Chemistry, University of Waterloo, 200 University Avenue West, Waterloo, ON N2L 3G1, Canada
Corresponding author:Karttunen, M.(mikko.karttunen@uwaterloo.ca)
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Abstract:The physical mechanisms behind hydrophobic hydration have been debated for over 65 years. Spectroscopic techniques have the ability to probe the dynamics of water in increasing detail, but many fundamental issues remain controversial. We have performed systematic first-principles ab initio Car-Parrinello molecular dynamics simulations over a broad temperature range and provide a detailed microscopic view on the dynamics of hydration water around a hydrophobic molecule, tetramethylurea. Our simulations provide a unifying view and resolve some of the controversies concerning femtosecond-infrared, THz-GHz dielectric relaxation, and nuclear magnetic resonance experiments and classical molecular dynamics simulations. Our computational results are in good quantitative agreement with experiments, and we provide a physical picture of the long-debated "iceberg" model; we show that the slow, long-time component is present within the hydration shell and that molecular jumps and over-coordination play important roles. We show that the structure and dynamics of hydration water around an organic molecule are non-uniform. © 2012 American Chemical Society.
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Controlled terms:Dynamics - Experiments - Hydrogen - Hydrophobicity - Molecular dynamics - Molecules - Sea ice
Uncontrolled terms:Ab initio - Broad temperature ranges - Car-Parrinello molecular dynamics simulations - Classical molecular dynamics - Computational results - Hydration shell - Hydration water - Hydrophobic hydration - Hydrophobic molecules - Long-time correlations - Microscopic views - Organic molecules - Physical mechanism - Physical pictures - Quantitative agreement -

Spectroscopic technique - Structure and dynamics - Tetramethylurea
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