

MAR14-2013-020222

Abstract for an Invited Paper
for the MAR14 Meeting of
the American Physical Society

Fast Quantum Molecular Dynamics Simulations of Shock-induced Chemistry in Organic Liquids

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The responses of liquid formic acid and phenylacetylene to shock compression have been investigated via quantum-based molecular dynamics simulations with the self-consistent tight-binding code LATTE. Microcanonical Born-Oppenheimer trajectories with precise conservation of the total energy were computed without relying on an iterative self-consistent field optimization of the electronic degrees of freedom at each time step via the Fast Quantum Mechanical Molecular Dynamics formalism [A. M. N. Niklasson and M. J. Cawkwell, Phys. Rev. B, **86**, 174308 (2012)]. The conservation of the total energy in our trajectories was pivotal for the capture of adiabatic shock heating as well as temperature changes arising from endo- or exothermic chemistry. Our self-consistent tight-binding parameterizations yielded very good predictions for the gas-phase geometries of formic acid and phenylacetylene molecules and the principal Hugoniot of the liquids. In accord with recent flyer-plate impact experiments, our simulations revealed i) that formic acid reacts at relatively low impact pressures but with no change in volume between products and reactants, and ii) a two-step polymerization process for phenylacetylene. Furthermore, the evolution of the HOMO-LUMO gap tracked on-the-fly during our simulations could be correlated with changes transient absorption measured during laser-driven shock compression experiments on these liquids.