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Modeling Photochemical Dynamics in Optically Active Energetic Materials

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— Most high explosives (HEs) absorb in the UV range, making it difficult to develop HEs that can be excited with standard lasers. The conventional optical initiation mechanisms require high laser intensity and occur via indirect thermal or shock processes. A photochemical initiation mechanism could allow control over the chemistry contributing to decomposition leading to initiation. We combine UV femtosecond transient absorption (TA) spectroscopy and excited state femtosecond stimulated Raman spectroscopy (FSRS) with Nonadiabatic Excited State Molecular Dynamics (NA-ESMD) to model the photochemical pathways in nitromethane (NM), a low sensitivity HE known to undergo UV photolysis. We investigate the ultrafast photodecomposition of NM from the $n\pi^*$ state excited at 266 nm. The FSRS photoproduct spectrum points to methyl nitrite formation as the dominant photoproduct. A total photolysis quantum yield of 0.27 and an $n\pi^*$ state lifetime of 20 fs were predicted from simulations. Predicted time scales reveal that NO_2 dissociation occurs in 81±4 fs and methyl nitrite formation is much slower at 452±9 fs corresponding to the absorption feature in the TA spectrum. The relative time scales are consistent with isomerization by NO_2 dissociation and ONO rebinding.

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