PROBING THE REACTION PATH OF $CH_3^+ + H_2 \rightarrow CH_5^+ \rightarrow CH_3^+ + H_2$ AND ISOTOPOLOGUES

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Protonated methane has long been of interest to astrochemists due to its presumed importance as a reaction intermediate in the reaction involving $CH_3^+$ + HD within the interstellar medium. Within the interstellar medium there is a nonstatistical H/D isotopic abundance observed for isotopologues of $CH_3^+$. While classical trajectory calculations have been performed dissociating $CH_5^+$ and $CH_4D^+$ into the fragments, $CH_3^+ + H_2$, $CH_2D^+ + H_2$ and $CH_3^+$ + HD[a], these calculations do not account for a large portion of the available energy being tied up in the zero point energy of the reactants and products. Earlier work in our group on $CH_2D_3^+$[b] showed the deuterium atoms were localized to the $CH_3^+$ group, rather than the $H_2$ moiety. Classical calculations fail to account for this observed localization, instead showing full delocalization of D between both $CH_3^+$ and $H_2$. With a quantum mechanical treatment, the energetics and wave functions will depend on which asymptotic channel is chosen, while in the classical treatment, these channels will be energetically equivalent. By performing Diffusion Monte Carlo simulations in Jacobi coordinates, we can constrain the distance between the $CH_3^+$ and $H_2$ subunits. Using this technique we have evaluated a one-dimensional reaction potential that includes the full anharmonic zero point energy in the remaining degrees of freedom and can determine how energetics of this reaction change upon partial deuteration of $CH_3^+$ or $H_2$. We have also evaluated the probability amplitude associated with the wave functions that are obtained in the DMC simulations at various values of the reaction coordinate.

[a] Z. Jin, B. J. Braams, J. M. Bowman, J. Phys. Chem. A 110, 1569 (2006).

[b] A. B. McCoy, B. J. Braams, A. Brown, X. Huang., Z. Jin, J. M. Bowman, J. Phys. Chem. A 108, 4991 (2004).