Investigation of Gas phase Decomposition Mechanisms in GaN-CVD by Theoretical Methods: The "Entropic Challenge"

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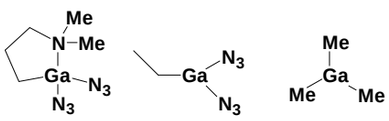
Initial results from density functional theory (DFT) calculations of the gas phase decomposition mechanisms of the classical $GaMe_3$ / NH_3 system and a single molecule galliumazide precursor (SMP) are presented. For the SMP system a plausible decomposition mechanism can be proposed. In general it is found that entropic contributions can compensate for bond energies, favoring radical pathways. Therefore, an accurate approximation of these finite temperature effects is absolutely crucial.

We report a survey of our first efforts to meet the "entropic challenge", trying to incorporate finite temperature effects and to differentiate between possible decomposition pathways by their relative reaction rates. We have focused on the MOCVD of GaN films both by the working Single Molecule Precursor (SMP) BAZIGA **1** and its model **2**, as well as the "classical" binary system $GaMe_3$ **3** plus NH_3. At this stage we have used approximate partition functions as opposed to a direct sampling of phase space via molecular dynamics. Possible deficiencies of this approach are discussed and in addition, the novel application of Variational Transition State Theory (VTST) to calculate the effective free enthalpy barrier of homolytic bond cleavage of **3** is shown. The main issue of this contribution are not so much the theoretical results, but to make the "entropic challenge" evident and to discuss the strategies to overcome the problem.



1 **2** **3**