

# Automated prediction of single bond activation pathways on small metal clusters: A case study of H<sub>2</sub> dissociation on Au<sub>n</sub> (n = 1 - 10)

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Chemical bond activation is the first important step in studies of chemical reaction mechanisms. Here, we take H<sub>2</sub> dissociation reactions on small gold cluster as an example. H<sub>2</sub> dissociation has attracted lots of interest because of its wide industrial applications. The number of isomers for chemical and physical adsorption of H<sub>2</sub> on gold clusters increases a lot with the increasing number of gold atoms. A good guess for TS or product is key point for obtaining important paths by traditional methods. It is difficult to get all the possible TSs and products only by conventional geometry optimizations. However, there is no guarantee to get the most important path even for small systems. Consequently, even for this simple single-bond activation reaction, there is still controversy about the factors that influence the catalytic activity of gold clusters.

In the present study, we made a thorough search for transition state (TS) structures for the H<sub>2</sub> dissociation channels on Au<sub>n</sub> clusters (n = 1 - 10), by using two recently-developed automated reaction path search methods [1-3], anharmonic downward distortion following (ADDF) and artificial force induced reaction (AFIR) methods. The search located numerous TS structures for the H<sub>2</sub> dissociation. Based on the extensive TS database, size effects, active sites, importance of metastable isomers, etc. are discussed comprehensively. Furthermore, we propose a general strategy for finding molecular dissociation channels on metal clusters automatically, on the basis of the two automated reaction path search methods. The calculation consists of the following three steps: (1) automated search for isomers of free gold cluster by ADDF method; (2) Search for reaction pathways between low-lying Au clusters obtained in step (1) and H<sub>2</sub> molecule by AFIR method; (3) refining the obtained AFIR paths to find TSs for H-H bond dissociation.

The calculated results show that the global minima are not always the best catalysts for H-H bond activation, and even for small gold clusters, some local minima should be considered. The activation barriers for H-H bond dissociation are compared for the energetically-favorable path and the path determined on the global minimum of gold clusters. The details will be shown in poster presentation.

In the present study, we only applied this strategy to the H-H bond activation. It can also be a promising tool to predict the best path for activation of other single bonds, such as C-H, C-C, etc.

## References

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