Development of Multi Component Molecular Theory and its Application

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Recently, our group has been developing some first-principles approaches which treat multi component systems including both electrons and nuclei (positron) quantum-mechanically: (I) Multi-component molecular orbital (MC_MO) [1] or density functional theory (MC_DFT) [2], (II) *ab initio* path integral molecular dynamics (PIMD) [3], and (III) multi-component quantum Monte Carlo (MC_QMC) [4] methods.

First, we will show the geometrical isotope effect (GIE) on $H_3O_2^-$ (Figure 1) with extensive range of temperature by PIMD method. At 50K the distributions of both H* for quantum $H_3O_2^-$ and T* for quantum $T_3O_2^-$ have single peak. The $\langle R_{OO} \rangle$ of H species is longer than that of T species, due to

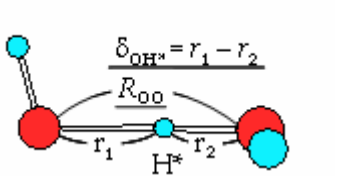

Figure 1. Schematic illustration of H_3O_2

the difference of zero point vibrational energy under anharmonic potential. At 400K the distribution of H* has single peak, while that of T* has double peaks. This means that the T* is difficult to go over the "effective potential barrier" with respect to proton transfer coordinate, since it becomes higher as the R_{OO} becomes longer. Actually, the $\langle R_{OO} \rangle$ of T species is longer than that of H species at high temperature, due to the multidimensional effect which is influence of both δ_{OH^*} and R_{OO} coordinates.

We will also show some recent results for "positronic compounds" by using (I) MC_MO or MC_DFT, and (III) MC_QMC methods.

References

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