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Grid calculation of reaction kinetic coefficients: a quantum mechanical approach

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Abstract

Introduction

Our communication provides a detailed description of the gridification of a quantum method for the direct calculation of kinetic coefficients by means of flux correlation functions and the Multiconfigurational time-dependent Hartree (MCTDH) scheme for the wavefunction. The methodology has been implemented and made available to the COMPCHEM VO within EGEE. The performance and realiability of the method is illustrated by presenting the results of a computational campaign aimed at the calculation of the N + N2 reaction rate coefficients using on the LAG3 potential energy surface.

Description

The direct calculation of chemical reactions thermal rates coefficients k(T) has increasingly attracted theoretical and computational work[1]. The computation of reliable thermal rate values can be of great use in several realistic multiscale simulations of complex Systems [2]. Whenever an elementary chemical reaction occurs through a reaction barrier and no long-living complex is formed, a rigorous method of calculating k(T) in a direct way. The method is based on a dynamics simulation confined in the region around the saddle point and has the advantage of decreasing significantly the numerical effort with respect to a full scattering simulation. The MCTDH scheme offers an additional numerical advantage to the approach [3] by expanding the time-dependent multidimensional wavefunction onto a basis of time-dependent functions.

Grid related relevance

It is accepted that the parametric nature of classical and semiclassical trajectory calculations makes these intrinsically suitable for implementation on computing grid and distributed computing naturally beneficial for related techniques. What is a new outcome of our work is that the flux correlation method can play an equivalent role for quantum calculations. This possibility is exploited by propagating separately the several quantum state basis functions. In this way, an increase of computational effort associated to the size of the full matrices to be propagated is transformed into a computational effort associated to the number of concurrent processes that need to be distributed.

This methodological advance exploits the availability of the computing grid by making of the innovative features of the EGEE available to the COMPCHEM VO.

References

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