Grid-Technology for Chemical Reactions Calculation

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Abstract. We discuss a possible strategy for implementing a grid-based approach to realizing the immense computational resources required to compute reactive molecular scattering cross sections and rate constants.

1 Introduction

Evaluation of chemical reaction cross-sections and rate-constants as well as more detailed scattering characteristics is one of the great challenges for computational technologies. The main difficulties are connected with the multi-dimension and multi-scale character of the problem. Both the accuracy and the computations of detailed state-to-state scattering cross sections are limited by the availability of computational resources and by the efficiency of the computational algorithm used in their calculation. We examine here a possible solution to these problems through the use of Grid technology. The main concept of Grid technology is the solution of a computational problem through *coordinated dynamic resource sharing within a multi-institutional virtual organizations* [1]. Some attempts at such Grid based solutions have already been implemented [2 - 4]. In these cases the Grid infrastructure supports the usage of standard quantum chemistry programs (Gaussian [5] and GAMESS [6]). This paper discusses a possible solution of the reactive scattering problem using a Grid based technology.

2 Description of Reactive Scattering Problem

We consider principally the process $A + (BC)_m \rightarrow (AB)_n + C$ which may take place with or without involving the formation of a quasi-bound or an activated complex $(ABC)^*$. When the process occurs via the formation of a quasi-bound complex, both the classical and quantum mechanical treatment of the process show chaotic behavior. The computation of the cross sections and rate constants require large, often unattainably large, amounts of computational resources on a conventional computer. Here we discuss an alternative approach that effectively utilizes Grid technologies.

The problem may be divided into two parts that may be solved separately. The first part being that of calculating the potential energy surface (PES) $V(q_1, q_2, \gamma)$ and the second part being the calculation of reaction probabilities and rate constants (or other properties). PESs are usually obtained from *ab initio* quantum chemical calculations (we consider these results as initial input to our problem), but they are represented in Cartesian co-ordinates that are usually not convenient for further calculations. For the scattering calculations the so-called curvilinear co-ordinates are the most convenient ones to use[7].

One of the computational problems consists in the transformation of the potential energy surface to the new co-ordinates $U(x^0, x^1, x^2) \equiv V(q_1, q_2, \gamma)$. It is also convenient to use certain analytic models for the PESs and to just determine their parameters. The PESs we develop depend on the fairly small number of parameters and these may be optimized to fit different sets of *ab initio* data [8]. In any case this problem may be solved separately. Since this problem is closely related to the inversion of a large dimensional matrix the corresponding methods may be used for its solution and these algorithms can also be parallelized. This procedure may easily be transferred to a Grid technology, implemented on a Grid of loosely connected computational systems. The calculation of scattering cross sections may be reduced to four sequential problems. The first one is the generation of the classical geodesic equations on the Lagrange surface of body system (at first this method of investigation classical dynamical systems was introduced by Krilov [9]) for the evolution of the system along the reaction coordinate and calculation of trajectory tube distribution; the second one is the problem of the solution Shroedinger equation on geodesic trajectory tubes; the third one is the calculation of the transition S -matrix elements and the fourth one is the calculation of transition probabilities [10, 11].

3 Grid for Chemical Reactions Calculation

The basic principles of the Grid for chemical reaction calculation can be stated as follows: 1) Present computer and data resources as a single virtual environment by developing a web portal on the Grid technology. 2) Build an easy-to-use user web interface for providing access to these resources. 3) Facilitate the sharing of results of research. 4) Organize archiving of input, output, and intermediate data.

As the basic software for Chemical Grid computing the Globus code will be used because of the following distinguishing advantages [12]. As the scheduling system we plan to use the Nimrod/G tool [13, 14], which is designed to manage the computational process including the transfer of input data and of the results of the calculations.

Returning to the problems arising in solving the reactive scattering problem that has been partly discussed in section 3, one must complete the following list of steps:

1) Computation of PES. 2) Parameterization of PES. 3) Solution of geodesic trajectory problems on Lagrange surfaces of the three-body system and the construction of the trajectory tubes distribution. 4) Calculation of the quantum system evolution on the trajectory tubes. 5) Computation of S -matrix elements and reaction probabilities for a set of initial phases φ_s and collision energies E_k^i . 6) Averaging of S -matrix element amplitudes over distribution of trajectory tubes, i.e. calculation of probabilities of elementary reactive quantum transitions. 7) Visualization of the results.

A Grid implementation of the solution of the reactive scattering problem implies the establishment of some infrastructure that should include algorithms for the *ab initio* calculation of PESs and the corresponding databases for different scattering partners. The next step then is the parameterization of the PESs using for instance the nonlinear optimization algorithm described in [9]. The Chemical Grid should contain a number of models that will permit the parameterizations of different aspects of PESs to varying accuracies. The classic geodesic trajectory problem calculation on the Lagrange surfaces for the reactive collision should be implemented for use in a distributed computational environment. Also the quantum reactive scattering part for the different trajectory tubes can be parallelized.

Our experience in complex problem solving using the Grid [15] will be used to achieve these goals. We wish to create a Chemical Grid that will allow the incorporation of both the data banks of different PESs and other properties accumulated by other scientists and our own computational codes.

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