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REGULAR ARTICLE

Quantum Wave Packet Dynamics of the N(^{2}D) +H_2 Reaction

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Abstract: In this work, the N+H_2 (v,j) reactions have been studied by means of quantum mechanical real wave packet method on an ab initio potential energy surface (PES) newly modified by Yang et al. [J. Phys. Chem. A 2013, 117, 3-8]. First accurate total and state-to-state reaction probabilities at total angular momentum J=0 have been calculated in a broad range of collision energies. Final state resolved reaction probabilities and product ro-vibrational distributions have been calculated at J=0. Initial state integral cross sections have been calculated by using Centrifugal Sudden approximation considering every total angular momentum without using any interpolation method. The effects of initial rotational and vibration on the dynamics have been also investigated. The obtained integral cross sections have been compared with the available data in the literature.

AMS subject classifications: 81U10, 81V45

KeyWords: State-to-state reaction probabilities, Centrifugal sudden, integral cross sections

Introduction

From atmospheric perspective, N+H_2 reaction is important to be understood the chemical processing of combustion systems that occurred in solar system, interstellar medium and molecular clouds. Especially, the compounds, involved metastable N(^{2}D) atom in these environments, are quite abundant and lead to various reactive scattering since the excited one

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is to be more reactive than the ground one[1-4]. The title reaction plays a central role in both experimental [5-6] and theoretical atmospheric point of view.

The first theoretical study on potential energy surface (PES) of title reaction was performed by H. Kobayashi and coworkers [7]. In that work, quasiclassical trajectory (QCT) studies were carried out to test of the potential energy surface. Then, Pederson et al. [8] reported the first global potential energy surface of this reaction that has been considered as prototypes of insertion reactions and has a deep potential well in transition state region with a small barrier in the entrance channel, on the contrary to the other insertion reactions[9]. Balucani and coworkers [1] implemented quantum mechanical, statistical and quasiclassical methods on the PES of Pederson [8]. Later on, Ho et al. improved the potential energy surface of Pederson and carried out QCT calculations [10]. There are many theoretical works in the literature on the N+H2 reactive scattering process [11-13].

The PES used in the present work, has been reported by Yang et al. [14] recently. This PES is based on an analytical function that gives more accurate results in the range of small internuclear distances when compared to the other previous surfaces mentioned above [14]. More detailed information for this PES can be found in ref. [14]. Since the NH_2 reaction, especially, has a deep well in the transition state region, NH_2 complex structure involves long-lived intermediate which tends to be an indirect reaction. In such reactions, the distributions of the internal quantum states of product molecule have been worth of being explored.

In this study, total and state-to-state accurate reaction probabilities, specific-initial and -final quantum state distributions of reactant and product are examined by means of a Real Wave Packet Method. This method is briefly outlined in section 2. The obtained results are discussed in part 3 of this text.

Theory

The method employed in this work is based on the use of real wave packet method, as originally introduced by Balint Kurti and Gray [15]. The time-dependent Schrödinger equation in Jacobi coordinates,

$$\hat{H}\psi(R,r,\gamma,t) = i\hbar\frac{\partial\psi(R,r,\gamma,t)}{\partial t} \tag{1}$$

The Hamiltonian for total angular momentum J=0 is expressed in terms of the Jacobi coordinates as

$$\hat{H} = -\frac{\hbar^2}{2\mu_R}\frac{\partial^2}{\partial R^2} - \frac{\hbar^2}{2\mu_r}\frac{\partial^2}{\partial r^2} - \frac{\hbar^2}{2}\left(\frac{1}{\mu_R R^2} + \frac{1}{\mu_r r^2}\right)\frac{1}{\sin\gamma}\frac{\partial}{\partial\gamma}\sin\gamma\frac{\partial}{\partial\gamma} + V(R,r,\gamma) \tag{2}$$