

Theoretical study of stereodynamics for the $S+H_2$ ($v=0-2, j=0$) \rightarrow $SH+H$ reaction

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Abstract. Quasi-classical trajectory (QCT) calculations for the reaction $S+H_2(v=0-2, j=0)\rightarrow SH+H$ have been performed in order to investigate the effect of initial vibrational states on both the scalar and vector properties. The integral cross sections, opacity function have been calculated. The results indicate that the reaction probability and the cross section increase as the initial vibrational quantum number increases. The vibrational distributions and rotational distributions at different vibrational excited states are presented, and the results are discussed in detail. In addition, the vector properties, involving scattering directions of reaction product and the alignment and orientation of rotational angular momentum, are calculated and discussed. The results indicate that the vibrational excited states have a positive effect on the forward scattering direction of the product, and the vibrational excited states have slight effect on the alignment and orientation of the rotational angular momentum.

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Key words: quasi-classical trajectory, integral cross section, ro-vibrational distribution, vector correlation.

1 Introduction

The combustion reaction of sulphur with hydrogen has attracted extensive attention because of its important role in the atmospheric chemistry and air contamination. The reaction $S+H_2$ is the simplest reaction involving the sulphur atom. The investigation on the reaction $S+H_2$ might reveal microscopic mechanism of $A+BC$ reaction. Therefore, both theoretical [1-9] and experimental [10, 11] studies on the title reaction have been reported.

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Maiti *et al.* [4] studied the intersystem crossing effect in the S+H₂ reaction by employing a “mixed” representation approach in combination with a trajectory surface-hopping method. Klos *et al.* [6] reported their theoretical study of the S(¹D)+H₂/D₂ → SH+H/SD+D, including the nonadiabatic effect. Berteloite *et al.* [7] performed kinetics and crossed-beam experiments in conditions approaching the cold energy regime, and Lee and Liu [10, 11] investigated the S(¹D) + H₂ reaction and its isotopic variants through Doppler-selected time-of-flight detection of the H or D product. An accurate *ab initio* potential energy surface (PES) for the lowest triplet state of H₂S was reported by Lv *et al.* [12], which makes it possible for us to study the vector properties of the reaction.

As mentioned above, most of the investigations on the reaction S+HH basically deal with the scalar properties. It is well known that the vector properties can reveal the details of the reaction with space information. To our best knowledge, only one study has been reported concerning the vector properties of the reaction S+H₂. Li *et al.* [13] investigated the effect of collision energy on the stereodynamics of the reaction S+HH/HD/DH/DD. They calculated the cross sections and vector properties at different collision energies. However the effect of vibrational excited states was not involved in their study.

In this paper, we calculated both the scalar and vector properties of the reaction S+H₂ at different initial vibrational excited states. The QCT method was present in Section 2; the result and discussion were present in Section 3; the conclusion was showed in Section 4.

2 Quasi-classical trajectory method

The quasi-classical trajectory (QCT) method employed in this study is the same as the method has been expressed particularly in previous works [14-26], we only present the calculation details of the current work. In our calculation, the classical Hamilton's equations are numerically integrated for motion in three dimensions, and the accuracy of the calculation is verified by checking the conservation of both the total energy and angular momentum. Batches of 100 000 trajectories have been run at each vibrational excited state. The integrate cross section (ICS) is calculated by $\sigma = \pi b_{\max}^2 p_r$, in which p_r represents the reaction probability, is given by N_r/N_t , where N_r and N_t represent the number of reaction trajectories and total trajectory number (100 000) respectively.

The center-of-mass (CM) frame [24-26] is chosen to describe the vector correlation, and its details are specified as follows. As can be seen in Fig. 1, k means the reactant relative velocity parallel to the z-axis, and k' is the product relative velocity. The x-z plane, containing vectors k and k' , is the scattering plane. θ_t represents the angle between k and k' , which indicates the scattering direction of the product. j' is the rotational angular momentum of product, whose polar and azimuthal angles are θ_r and ϕ_r , respectively. $P(\theta_r)$ and $P(\phi_r)$ describe the probability density distribution of reaction products, reflecting $k-j'$ and $k-k'-j'$ vector-correlation respectively. The alignment and orientation of j' can be obtained by analyzing and $P(\theta_r)$ and $P(\phi_r)$ distributions. Four generalized