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# Theoretical study of the stereo-dynamics of the reaction H+CH→ $H_2$+C($^1$D)

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**Abstract.** The vector correlations between products and reagents for the reactions H+CH→ $H_2$+C($^1$D) at different collision energy have been studied by using the quasi-classical trajectories (QCT) on an ab initio potential energy surface of 1A′ symmetry. Four polarization-dependent generalized differential cross-sections (PDDCSs) have been calculated in center-of-mass (CM) frame. The distribution $P(\theta_r)$ of the angle between $k$ and $j'$, the distribution $P(\phi_r)$ of dihedral angle denoting $k-k'-j'$ correlation in the form of polar plots are calculated as well. The effect of deep well in potential surface and differ collision energies on the alignment and the orientation of product molecule $H_2$ rotational angular momentum vectors $j'$ is revealed.

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**Key words**: quasi-classical trajectories, vectors correlations, stereodynamics

## 1 Introduction

In the recent decade, the $CH_2$ reactive system have been received considerable attention because of their importance in combustion reactions[1], gasification of coal[2], and astrochemistry[3]. For instance, the generation of methylene radical $CH_2$ by the title reaction is a key parameter in the chemistry of hydrocarbon combustion. From a practical perspective, $CH_2$ reactive system has important implications in organic chemistry, particularly in understanding the mechanisms of a large number of carbon-alkane reactions. Much experiment have focused on the C($^1$D)+ $H_2$ → CH+H reaction[4-9]. In recent years, Bergeat *et al.* measured the product angular and time-of -flight (TOF) distribution for the C(1D)+ $H_2$ reaction using the crossed molecular beam (CMB) experiment[10]. Balucani *et al.* obtained the product angular and velocity distributions in crossed beam experiments[11].

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Theoretically, $CH_2$ reactive system contains only eight electrons and is thus amenable to high level ab initio calculations of its potential energy surface. Indeed, a highly accurate global potential energy surface of the 1A′state has been reported by Bussery-Honvault *et al.*[12]. They constructed the PES of $CH_2$ system and calculated the reaction probabilities for the reaction $C(^1D)+ H_2$ and present that the state-to state reaction probabilities as a function of the collision energy show a dense resonance structure, which is the first time reported for this type of atom + diatom reaction. Subsequently, the availability of the high quality potential energy surface has stimulated several dynamical studies of the reaction $C(^1D)+H_2$. Balucani *et al.*[13] performed the quantum mechanical (QM) scattering calculations and quasi-classical trajectory (QCT) calculations for the translational energy distributions. Banares *et al.* calculated integral and differential cross sections at 80meV collision energy and they also obtained the total and vibrationally state-resolved reaction probabilities in the 0-0.5eV collision energy range. Later, wave packet studies for integral cross sections and rate constants were reported by Lin and Guo[14,15].

As mentioned above, most of the studies so far have focused on the $C(^1D)+H_2 \rightarrow CH+H$ reaction. Because the importance in atmospheric and combustion chemistry, we also should pay attention to the reverse reaction $H+CH \rightarrow H_2+C(^1D)$. However, we have not found any experimentally nor theoretically research about the reverse reaction. This work is the first example for reverse reaction $H+CH \rightarrow H_2+C(^1D)$. Although the QCT method yields important information with relatively low computational costs, the results may be plagued by the neglect of quantum effects such as zero-point energy and tunneling. These difficulties have until now prevented us from an accurate result of scalar properties, such as reaction probability, reactive cross sections, thermal rate coefficient. However, vector properties (such as velocities and angular momentum) based on QCT calculation will be more accurate. In order to understand the dynamics of the H+CH reaction completely, it is necessary to study its vector properties, which can provide more information about chemical reaction stereo-dynamics [16-30]. By comprehending the scalar and vector properties together, the full picture of the scattering dynamics can be presented.

## 2 Quasi-classical trajectory calculations

In this paper we only summarize the details relevant to the present work. In the center-of-mass (CM) frame shown in Fig. 1, the reagent relative velocity vector $k$ is parallel to the z-axis. The $x-z$ plane is the scattering plane containing the initial and final relative velocity vectors, $k$ and $k'$. $\theta_t$ is the angle between the reagent relative velocity and product relative velocity, namely, scattering angle. The angles $\theta_r$ and $\phi_r$ are the polar and azimuthal angles of the final rotational momentum $j'$. The distribution function $P(\theta_r)$ describing the $k-k'$ correlation can be expanded in a series of Legendre polynomials [24] and the dihedral angle distribution function $P(\phi_r)$ describing the $k-k'-j'$ correlation expanded in a Fourier series. The full three-dimensional angular distribution associated