Quasiclassical trajectory study of the stereodynamics for the Au + H$_2$ (v=0, j=0) → AuH + H reaction

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Abstract. The stereodynamics of the reaction Au+H$_2$ have been performed by using quasi-classical trajectory (QCT) method on a global potential energy surface created by Zanchet *et al.* at the collision energy of 1.8, 2.2, 3.0 eV. The calculation on $P(\theta_r)$, $P(\phi_r)$, and $P(\theta_r,\phi_r)$ and four polarization-dependent differential cross sections (PDDCSs) in the center-of-mass (CM) indicate the dependence of the product polarization on collision energies. The product rotational angular momentum vector j' is symmetric and perpendicular to k direction according to the $P(\theta_r)$ distributions. The $P(\phi_r)$ distributions around ϕ_r=270° indicate that the rotational angular momentum vectors not only aligned along the y-axis direction, but also oriented to the y-axis negative direction. For the lower collision energy of 1.8eV, the PDDCS$_{00}$ is symmetric on the forward and backward scattering, probably due to the long lifetime complex created in the insertion reaction, while for the increasing collision energies, the prominent forward scattering is all on account of that the reaction is controlled by direct stripping.

1 Introduction

The point of view that Au is inactive as catalysts was changed by Haruta's discovery [1, 2]. Since then, Au catalysis has attracted much interest for many reactions, including low-temperature CO and alcohol oxidation [3-7], water-gas shift [8], selective hydrogenations [9-17]. This renaissance originates from the application of Au cluster catalysts which are

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prepared by innovative methods and exhibit unique reactivity different from the bulk Au catalysts.

Although the Au catalyzed hydrogenation reactions are less intensively investigated than the oxidation reaction, there are theoretical and experimental studies on the hydrogenation reactivity. Stobinski and Claus proposed that H_2 reacts and dissociates on low coordinated Au atoms at the corners or edges of the clusters [13-17]. Corma *et al.* indicated that Au atoms, which locate not only at corner or edge low coordinated positions but also not directly bonded to the supports, are active for H2 dissociation by means of periodic DF calculations [18].

The reactivity of Au cluster correlates closely with the structural and electronic properties of the cluster [19-22]. The ground atom structure $5d^{10}6s^1$ exhibits the strong relativistic effect which makes a reduced 5d-6s energy gap and strong s-d hybridization [23], as the result, the structures of Au cluster present variedly, linear (one-dimension) [19], planar (two- dimension) [24,25], and shell-like (three dimension) [26]. Recently, Zanchet *et al.* made a series of studies about structure, charge effects for the H_2 dissociation on Au cluster and obtained very interesting results [27-30]. H_2 dissociation on the linear gold chains with no barrier along the minimum energy path, and higher barriers presented on the planar clusters, however, the increased reactivity of the folded planar Au cluster is originated from the orbital flexibility when the s-d hybridization is broken. Moreover, Zanchet *et al.* established a global potential energy surface (PES) for title reaction by fitting highly correlated *Ab initio* method which is less adapted when the number of involving electrons is large. They performed quantum wave pocket and quasiclassical trajectory (QCT) calculation using the PES, and proposed that two different mechanisms control the reaction, direct and indirect. In indirect reaction mechanism, dominating at low collision energies, presence of insertion well, which is similar with the deeper chemisorption well of larger gold clusters reacting with H_2, stems from conical intersections and curve crossings with the excited electronic states Au (^{2}D) and Au (^{2}P) [28].

However, previous works mainly deal with scalar properties, vector properties representing stereodynamics of chemical reactions are not performed. The stereodynamic properties such as velocities and angular momentum with translational and rotational energies are investigated not only by magnitudes but also by well-defined directions [31-36]. Analyzing reaction dynamics together scalar with vector can provide a complete understanding on reaction dynamics. However, up to now, no literature reported the vector properties about the reaction. In order to fully understand the reaction, we investigated the three angular distributions of $P(\theta_r)$, $P(\phi_r)$, and $P(\theta_r, \phi_r)$ and four polarization-dependent differential cross sections (PDDCSs) based on the PES from Zanchet *et al.* [28].