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Formation of NaH Molecules in the Lowest Rovibrational Level of the Ground Electronic State via Short-Range Photoassociation

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Abstract. The formation of NaH molecules in the lowest rovibrational level of the ground electronic state is investigated using a pump-dump photoassociation (PA) scheme. In short-range region, two colliding atoms Na and H are efficiently associated into the NaH molecule in the rovibrational $|0,0\rangle$ state of the ground electronic state via the intermediately rovibrational $|10,1\rangle$ state of the excited electronic state. The changes of populations with the electric field amplitudes, frequency detunings, dump pulse duration and delay time between two laser pulses are calculated and discussed. The PA probability reaches 0.623 with a high state-selectivity.

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Key words: Short-range photoassociation, NaH molecule, rovibrational state.

1 Introduction

In recent years, the control of atomic and molecular processes with ultrashort laser pulses has made considerable progress, including photoassociation (PA) [1–4], photodissociation [5–10] and laser-control of molecular orientation [11–15]. In a PA process, external laser field induces colliding atoms to associate into molecules. The PA process of cold ($T < 1$ K) or ultracold ($T < 1$ mK) alkali-metal atoms taken place in a long internuclear range ($2 < R < 20000$ a_0, where a_0 is Bohr radius) has recently been widely investigated by using the shaped and chirpped laser pulse techniques [16-22]. And a small number of research works have focused on the short-range ($R < 30$ a_0) PA process of atoms at high temperature ($T > 1$ K) [23–32]. The short-range PA at high temperature

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is a more challenging and significance research topic in physics and chemistry. Engel and co-workers studied the PA reaction driven by the shaped laser pulse using the local control theory [23, 24]. Kosloff and co-workers focused their attention on the PA process of thermally hot atoms [25, 26]. Korolkov *et al*. investigated theoretically the vibrational sate-selectivity PA reaction [27]. de Lima *et al*. calculated the PA probability using one-dimensional wave-packet method [28, 29]. We investigated theoretically the PA and photodissociation processes driven by the femtosecond and picosecond laser pulses [30–32].

How to produce stable photoassociated molecules is an important topic in the quantum control research field [24, 33], since the PA molecules in the lowest rovibrational level of the ground electronic state are the most stable ones. Up to now, most of theoretical schemes for PA processes only study the control of vibrational states, and not take rotational states into account. In the present work, we study the short-range PA process to prepare the NaH molecule in the lowest rovibrational state of the ground electronic state by using a pump-dump PA scheme including two electronic states and the vibrational and rotational degrees of freedom. The numerical simulation shows that a high PA yield can be achieved using this simple PA scheme.

2 Theoretical calculation methods

In the PA process of Na and H atoms, the Hamiltonian describing the transition from the ground $X^1\Sigma^+$ electronic state to the excited $A^1\Sigma^+$ electronic state can be expressed as

$$\hat{\mathbf{H}}(t)=\begin{pmatrix} \hat{T}(R,\theta)+V_g(R)+W_{11}(R,\theta,t) & W_{12}(R,\theta,t) \\ W_{21}(R,\theta,t) & \hat{T}(R,\theta)+V_e(R)+W_{22}(R,\theta,t) \end{pmatrix}, \tag{2.1}$$

where R is the internuclear distance and θ the angle between the laser polarization and the molecular axis. $V_g(R)$ and $V_e(R)$ denote the potentials of the ground $X^1\Sigma^+$ and excited $A^1\Sigma^+$ electronic states, respectively. The kinetic energy operator $\hat{T}(R,\theta)$ is given by

$$\hat{T}(R,\theta)=\hat{T}_k+\hat{T}_\theta \tag{2.2}$$

with

$$\hat{T}_k=-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial R^2} \tag{2.3}$$

and

$$\hat{T}_\theta=-\frac{\hbar^2}{2mR^2}\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\Big(\sin\theta\frac{\partial}{\partial\theta}\Big), \tag{2.4}$$

where m is the reduced mass of the NaH molecule. The laser-molecule interaction Hamiltonian $W_{ij}(R,\theta,t)$ is expressed as

$$W_{ij}(R,\theta,t)=-\mu_{i,j}(R)\varepsilon(t)\cos\theta, \tag{2.5}$$