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The continuous-time quantum walk with transition moments of the Gaussian distribution as a mathematical model of rotational excitation in molecules by an optical pulse

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Abstract. The time evolution of the rotational distribution of diatomic molecules induced by a train of broadband optical pulses can be modeled as a class of continuous-time quantum walk (CTQW) on a half line. As the transition moments asymptotically decrease to zero like Gaussian distribution, the rotational distribution cannot reach infinity far from the initial state and exhibits a localization-like behavior where the upper limit of the distribution is not trivial. We investigated a time evolution of the density distribution with the transition moments of the Gaussian distribution peaked at the boundary of the half line for the initial state of $J=0$. Even though the time evolution exhibited oscillatory motion inside a certain region repeatedly, we observed a clear time-averaged distribution that is well fitted to the stretched exponential distribution with parameters close to the Gaussian distribution by numerical simulation. By varying the coefficients of the Gaussian distribution of the transition moments, the coefficients of the stretched exponential distribution can be determined by regression.

1. Introduction

Quantum mechanics of a rotator has been of interest for mathematics and physics for many decades, thereby constructing a foundation of atomic or molecular physics. The quantum kicked rotor (QKR) is one of the standard models, which describe the development of angular momentum by periodical kicking of delta pulses[1]. Recently, the dynamics of the QKR was experimentally implemented on cold atoms[2, 3, 4] and diatomic molecules[5, 6]. The characteristics of the QKR was studied on real molecules, such as the so-called quantum resonance[7] and the localization similar to Anderson localization or Bloch oscillation[8, 9]. Currently, the series of rotational states in diatomic molecules is considered as a one-dimensional network of quantum states connected by optical transition, which provides a testing field of fundamental quantum transport dynamics. The characterization of localization in a broad sense is also important for practical applications such as isotope separation by molecular selective excitation by using a coherent light source[7, 10].

The time evolution of the rotational distribution in the QKR can be described as a class of continuous-time quantum walk (CTQW)[11]. The CTQW is a well-studied mathematical
model[12, 13, 14]. To develop simple mathematical models of the localization in the QKR from the viewpoint of the CTQW, we classify the localization into four types and attempt to characterize each type of localization independently[15]. We derived an empirical unified parameter to characterize the range of localization for the localizations, namely interval mismatch (Type 1) and centrifugal distortion (Type 2), caused by rotational coherence[16]. The remaining types are the lack of spectral amplitude (Type 3) and the augmentation of spectral amplitude (Type 4), which are caused by the relative distribution of transition moments. The CTQW with the transition moments of the Gaussian distribution is a practical model to investigate Type 3 independently, because the distribution of the spectral amplitude of optical pulses are frequently approximated by Gaussian functions. However, it is not discussed explicitly, because the independent implementation of such a process is difficult in real molecules.

In this paper, we discuss the asymptotic behavior of a density distribution on the CTQW on a half line with the transition moments of the Gaussian distribution. The model emulates the time evolution of the rotational distribution with a broadband optical pulse where other effects, such as multiphoton processes or interferences relevant to the rotational coherence are neglected. To develop a foundation of mathematical physics of the localization of the rotational distribution in molecules, we performed the numerical analysis of the simplest case in symmetry.

2. The model and method

The modeling of molecular rotational excitation by optical pulses was shown in our previous studies[16, 17]. By neglecting the effect of pulse–interval mismatch and centrifugal distortions, the time-dependent Schrödinger equation system for molecular rotational distribution in optical pulses can be expressed as follows:

$$-i\frac{d}{dt}C_J(t) = \sqrt{\omega_{J-1}}C_{J-1}(t) + \sqrt{\omega_J}C_{J+1}(t),$$

where $C_J(t)$ is the complex amplitude of the rotational states $J$ at time $t$, $\sqrt{\omega_{J-1}}$ is the transition moment from $J$ to $J + 1$. The probability density distribution of state $J$ is given by $|C_J(t)|^2$. The transition moments of the Gaussian distribution peaked at the boundary of a half line can be expressed as follows:

$$\sqrt{\omega_J} = \begin{cases} 0 & (J < 0) \\ A_g \exp \left(-\left(\frac{\log 2}{H_g}\right)^2\right) & (J \geq 0) \end{cases},$$

where $H_g$ is the half width half maximum (HWHM) of the Gaussian distribution. The parameter $A_g$ is fixed at 1.0. This parameter makes no influence on the time evolution except the speed of the evolution. The initial state is fixed as follows:

$$C_J(0) = \delta_{J,J_0}.$$

In this study, we considered only $J_0 = 0$.

The time evolution was evaluated by solving Eqs.(1–3) numerically using the fourth-order Runge–Kutta method. The parameter $H_g$ was varied from 5.0 to 50.0 for regression. Some examples of the distribution of the transition moments are shown in Figure 1. The number of $J$ states considered in the calculation was $10H_g$, which was sufficient to ignore the influence of the upper boundary of the system. The evolution was simulated till $t_{\text{end}} = 500H_g$ by a step of 0.025. The numerical error estimated from the sum of probability distribution was in the order of $10^{-5}$. The time-averaged distribution was obtained by using the following approximation:

$$\rho(J) = \lim_{t \to \infty} \frac{\int_0^t |C_J(t)|^2 dt}{t} \approx \frac{\sum_{t=0}^{t_{\text{end}}} |C_J(t)|^2}{t_{\text{end}}},$$

where only integer $t$ was included in the summation.
3. Results and Discussion
We executed the computer simulations of the model described by Eq.(1). The examples of the transition moments that were used in simulations are shown in Figure 1. The result of time evolution of probability distribution obtained by the simulation with \(H_g=20\) is shown in Figure 2. Note that this figure shows only the first one-tenth of the simulation. Around \(t=0\), the population appears as if moving toward the high \(J\) states at a nearly linear propagation speed. However, a massive reconnection behavior is observed around \(J=20\). One of the primary parts of the population approaches the region around \(J=60\) while producing many branching reflections. The entire time evolution shows interference-like patterns caused by many reconnection waves. The upper limit of the propagation observed in the linear scale is about \(J=60\), which is a position similar to one where the relative transition moment approaches zero. The characteristics observed here were common regardless of input \(H_g\) except for the width of the distribution.

![Figure 1](image1.png)  
**Figure 1.** Examples of Gaussian distribution of relative transition moments obtained with \(H_g=10, 20,\) and 40.

![Figure 2](image2.png)  
**Figure 2.** Example of time evolution of \(|C_J(t)|^2\) simulated with \(H_g = 20.0\) and \(J_0 = 0\).

![Figure 3](image3.png)  
**Figure 3.** Examples of time-averaged distribution \(\rho(J)\) simulated with \(H_g=10, 20,\) and 40.

![Figure 4](image4.png)  
**Figure 4.** Coefficients of time-averaged distributions simulated with various \(H_g\) fitted by stretched exponential function.
Figure 3 shows the examples of time-averaged distribution $\rho(J)$ obtained by simulation. Even though the time evolutions were oscillatory, smooth structures were observed. The distributions comprise two peaks. One is the sharp peak at $J=1$, and the other is the broad peak with properties similar to the Gaussian distribution around the maximum. A small interference pattern was observed between the two peaks, which is also a common characteristic.

We focus on the shape of the broad peak, because it determines the upper limit of the propagation. We fitted the broad peak by using the stretched exponential function given by

$$f(J) = A_e \exp \left[-\left(\frac{\log 2}{H_e} (J - x_e)\right)^{\gamma_e}\right],$$

where $H_e$ provides the HWHM for any $\gamma_e$. $f(J)$ provides the Gaussian distribution for $\gamma_e=2.0$. $A_e$ is not very important, because it is nearly the same as the normalization factor.

Figure 4 shows the coefficients of the time-averaged distribution fitted by Eq. (5). The fitting errors were not more than 0.1, which are not displayed except $\gamma_e$ for small $H_g$. A linear scaling rule for $H_g$ was observed for $H_e$ and $x_e$. We arrived at the following relation by regression of the obtained data:

$$H_e = A_1 H_g + B_1 \quad (A_1 = 0.904 \pm 0.006, B_1 = -0.132 \pm 0.177),$$

$$x_e = A_2 H_g + B_2 \quad (A_2 = 0.572 \pm 0.005, B_2 = 1.020 \pm 0.152).$$

By considering the coefficients and the error obtained, we propose a hypothesis where the coefficients can be expressed in a simple analytical form:

$$H_e \simeq \frac{2}{\pi \log 2} H_g, \quad x_e \simeq \frac{1}{\sqrt{\pi}} H_g + 1. \quad (8)$$

Almost no dependence was observed on $H_g$ for $\gamma_e$. The average value was $\gamma_e=1.922 \pm 0.008$, which was evidently less than $\gamma_e=2.0$ of the Gaussian distribution. We observed that the stretched exponential function is essential to describe the distribution analytically. However, the Gaussian distribution still seems to be a good approximation function for expressing the distributions, because it is in good agreement with the obtained distribution.

4. Conclusion

We performed numerical analysis of a simple mathematical model of the CTQW with the transition moments of the Gaussian distribution, which describes one of the characteristics of the molecular rotational excitation by an optical pulse. We observed smooth time-averaged distributions that can be fitted by the stretched exponential function. The order of the stretched exponential $\gamma_e$ was close to the Gaussian distribution. However, the evident difference from the Gaussian was evaluated numerically. The coefficients obtained from regression were analytically expressed. However, those should be validated by mathematical analysis using approximate methods, because the model is not solvable. The upper limit of the population propagation in linear scale was not considerably different from the intuitive estimation from the distribution of the relative transition moment. However, the shape of the time-averaged distribution had not been known as far as authors’ knowledge, although this model expresses fundamental characteristics of population transfer by optical transitions.

The peak position of the Gaussian distribution and the initial state should be varied to obtain the general tendency of the asymptotic behavior of this model. In fact, the case investigated in this study was a special case from the viewpoint of the symmetry. Moreover, the interaction between other localizations should be investigated for application in real molecules. In particular, the interaction between the localizations induced by the centrifugal distortion is of interest, because it exhibits nearly the same range of localization as the time-averaged distribution obtained in this study.
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