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2006 J. Phys. B: At. Mol. Opt. Phys. 39 421

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Rotational entangled states between two coupled molecules

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Received 29 August 2005, in final form 30 November 2005

Published 19 December 2005

Online at stacks.iop.org/JPhysB/39/421

Abstract

A method is proposed to create entanglement between two coupled identical polar molecules separated at a distance of tens of nanometres. Entangled states of two coupled polar molecules controlled by half-cycle pulses are studied theoretically. The von Neumann entropy is calculated to characterize the degree of entanglement. By varying the pulse shape of the applied laser, transition from regular to irregular behaviour occurs. Moreover, the entanglement is also found to be enhanced by applying multiple laser pulses. The behaviour from quantum to classical limit is also discussed.

1. Introduction

The control of the molecular alignment and orientation [1] is important in stereodynamics, trapping molecules, molecular focusing, surface catalysis and nanoscale design. The alignment is due to the anisotropic polarizability induced by nonresonant pulses. The pendular states—hybrid of field-free molecular eigenstates [2–4]—can be created by turning on a picosecond laser pulse adiabatically. For polar molecules, a femtosecond laser pulse is able to generate a field-free orientation [5–7]. Experimentally, several methods concerning femtosecond photodissociation spectroscopy and ion imaging have been employed to measure the rotation of molecules [8–14].

Recently, the coupled-rotor model has attracted much interest [15, 16] because some physical properties such as dielectric response may display peculiar behaviour in the presence of dipole–dipole interaction [17]. In some materials, molecules are found to show a free rotation. For example, NH₃ groups behave like one-dimensional quantum rotors in certain Hofmann clathrates [18]. In particular, a line broadening mechanism is proposed based on rotor–rotor coupling. With the advances of nanotechnology, one can investigate the quantum rotors which are mounted on the surfaces [19–21]. From the laser spectroscopy, two individual fluorescent molecules separated by several nanometres on the surface of an organic crystal can be resolved. The coherent interactions between the dipole moments associated with their

optical transitions are found in the quantum optical measurements. The strong dipole–dipole coupling produces entangled subradiant and superradiant states in the two molecules system under laser radiation [19].

Much research has been devoted to generating entanglement in quantum-optic and atomic systems [22, 23] owing to its essential importance in quantum information processing [24, 25]. For molecular systems, most of the works are focused on the implementations of quantum computation [26]. In general, the qubits are encoded by the vibrational (rotational) modes of the molecules [27, 28]. In order to realize the quantum logic gates experimentally, the optimal control theory is usually applied to design the suitable laser pulses. However, studies on the entanglement of coupled molecules under strong laser fields still receive little attention so far [29], especially for a system evolving from a non-entangled case to an entangled one. Although some similar studies have been investigated, these works are limited in the model of kicked tops [15, 16]. In this paper, we consider a more realistic system for this physical interest. A method is proposed to create entanglement between two coupled identical polar molecules separated in a distance of tens of nanometres. Both molecules are assumed to be irradiated simultaneously by the laser pulses. It is found that the entanglement induced by the dipole interaction can be enhanced by applying single or multiple laser pulses. Moreover, the asymmetrical laser shape is also found to influence the degree of the entanglement. The crossover from quantum to classical limit is also discussed by varying the Planck constant.

2. Model

Consider now two diatomic polar molecules (e.g. NaI) separated in a distance of R . The molecule system is irradiated by laser pulses. The total Hamiltonian can be written as

$$H = \sum_{j=1,2} \frac{\hbar^2}{2I} L_j^2 + U_{\text{dip}} + H_{\text{f-m}}, \quad (1)$$

where L_j^2 and $\frac{\hbar^2}{2I}(=B)$ are the angular momentum operator and rotational constant, respectively. U_{dip} is the dipole interaction between two molecules:

$$U_{\text{dip}} = \frac{[\vec{\mu}_1 \cdot \vec{\mu}_2 - 3(\vec{\mu}_1 \cdot \hat{e}_R)(\vec{\mu}_2 \cdot \hat{e}_R)]}{R^3}, \quad (2)$$

where $\vec{\mu}_1$ and $\vec{\mu}_2$ are the dipole moments. The dipole moments of two molecules are assumed, for simplicity, to be identical, i.e. $\mu_1 = \mu_2 = \mu$. The field–molecule coupling $H_{\text{f-m}}$ can thus be expressed as

$$H_{\text{f-m}} = - \sum_{i=1,2} \mu E(t, \nu) \cos \theta_i, \quad (3)$$

where θ_1 and θ_2 are angles between dipole moments and laser field. The laser field is given by $E(t, \nu) = E_0 f(t) \cos(2\pi \nu t)$, where E_0 is the field strength and ν is the frequency. The envelope function $f(t)$ is assumed to be Gaussian shape centred at the time $t = t_0$ with duration σ , i.e. $f(t) = e^{-(t-t_0)^2/\sigma^2}$. Traditionally, a half-cycle pulse (HCP) is a strongly asymmetric monocycle pulse that consists of two parts: a very short, strong pulse and a much long and weak tail of opposite electrical field. The pulses $E(t, \nu)$ used in the present work are actually not the exact half-cycle pulses as defined in [30]. However, practical calculation shows that there is almost no influence on our final result if a long and weak tail is introduced in the pulses $E(t, \nu) = E_0 f(t) \cos(2\pi \nu t)$. Thus, it is reasonable to model a half-cycle pulse by using the function $E(t, \nu)$ in our calculation. In addition, the field duration is considered to be much shorter than the molecular rotational period in our work. Based on these conditions,

an impulsive model can be employed in this case [6, 31]. The time-dependent Schrödinger equation can be solved by expanding the wavefunction Ψ in terms of a series of field-free spherical harmonic functions $Y_{lm}(\theta, \phi)$ as

$$|\Psi\rangle = \sum_{l_1 m_1 l_2 m_2} c_{l_1 m_1 l_2 m_2}(t) |Y_{l_1 m_1}(\theta_1, \phi_1)\rangle |Y_{l_2 m_2}(\theta_2, \phi_2)\rangle, \quad (4)$$

where (θ_1, ϕ_1) and (θ_2, ϕ_2) are the coordinates of the first and second molecules, respectively. The time-dependent coefficients $c_{l_1 m_1 l_2 m_2}(t)$ correspond to the quantum numbers $(l_1, m_1; l_2, m_2)$ and can be determined by solving Schrödinger equations numerically. In equation (4), the inter-molecule separation R is assumed to be fixed for simplicity, so that the total wavefunction has no spatial dependence. Although the variation of R might be inevitable due to the influence of laser fields or inter-molecule vibrations, however, recent experiments exhibited that the spatial resolution in tens of nanometres for two individual molecules hindered on a surface is practically possible [19–21]. In principle, the free orientation model can be easily generalized to the hindered ones by replacing the spherical harmonic functions with hindered wavefunctions as shown in our previous work [32]. As shown in [32], the essential physics for both cases are very similar. Thus, the discussion in the free orientation model should be meaningful.

Let us now focus on the entanglement generated in our system. The coupled molecules can be expressed as a pure bipartite system: $|\Psi\rangle = \sum_{l_1 m_1 l_2 m_2} c_{l_1 m_1 l_2 m_2}(t) |Y_{l_1 m_1}\rangle |Y_{l_2 m_2}\rangle$. The *reduced* density operator for the first molecule is defined as

$$\rho_{\text{mol}1} = \text{Tr}_{\text{mol}2} |\Psi\rangle\langle\Psi|. \quad (5)$$

To study the degree of entanglement, the bases of molecule 1 are transformed to make the reduced density matrix $\rho_{\text{mol}1}$ diagonal. The entangled state can be represented by a biorthogonal expression with positive real coefficients λ_{lm} which can be obtained by diagonalization of the density matrix $\rho_{\text{mol}1}$. The degree of entanglement for the coupled molecules can then be measured by von Neumann entropy [33, 34]

$$\text{entropy} = - \sum_{lm} \lambda_{lm} \log_n \lambda_{lm}. \quad (6)$$

In our work, NaI molecule in the ground state with dipole moment 9.2 debyes and rotational constant 0.12 cm^{-1} is used. The duration and centre of the pulse are set to 279 fs and 1200 fs, respectively. The crossover from the non-entangled case to the entangled one is studied based on the initial condition $c_{0000}(t=0) = 1$.

3. Results and discussion

Figure 1 illustrates the time evolution of the entropy with different ratios in magnitude of the positive and negative peak values of the laser pulse as R is set equal to $1.5 \times 10^{-8} \text{ m}$. The laser frequency is tuned to change the ratio as shown in the inset by fixing other parameters. For the case of ratio 9:1, an irregular-like behaviour is obtained with time-averaged value 0.51. If the ratio is set equal to 1:1, the entanglement shows a nearly periodic behaviour with small averaged entropy. This result is very similar to the limiting case without laser, and indicates that the entanglement depends sensitively on the ratio of the laser pulse, i.e. the excitation is suppressed under the condition of 1:1 ratio. Meanwhile, the dipole force only establishes periodic-like entropy.

Let us consider the first ten most contributive coefficients λ_{lm} . The λ_{lm} are re-arranged and denoted as λ_p with $p = 1, 2, 3, \dots$. For example, λ_1 is the most contributive coefficient. The insets of figure 1 show the ten coefficients (λ_p) at short and long-time regimes. In the case of

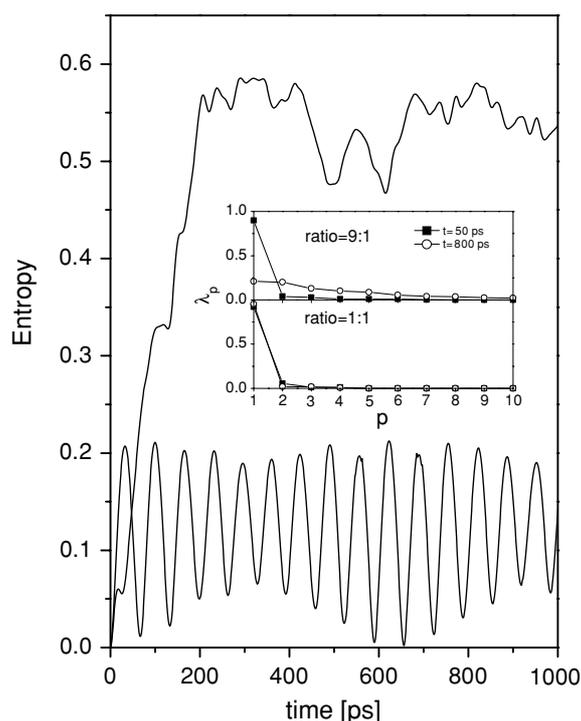


Figure 1. Time evolution of the entropy after applying single laser pulse for different ratios in magnitudes of the positive and negative peak values of the laser pulse. The graphs show the irregular (periodic) behaviour for ratio 9:1 (1:1), corresponding to the frequency $8.6 \times 10^{11} \text{ s}^{-1}$ ($1 \times 10^{12} \text{ s}^{-1}$). Inset: the first ten contributive coefficients λ_{Im} are arranged and denoted by λ_p at short time ($t = 50 \text{ ps}$) and long time ($t = 800 \text{ ps}$), corresponding to the cases of ratios 9:1 and 1:1, respectively. The field strength and inter-molecule separation are $E_0 = 3 \times 10^7 \text{ V m}^{-1}$ and $R = 1.5 \times 10^{-8} \text{ m}$.

the ratio 9:1, the eigenvalue λ_1 dominates the contributions at short-time regime ($t = 50 \text{ ps}$). However, the contributions are distributed more averagely between different levels as $t = 800 \text{ ps}$ regime. This means the system is in some sort of dynamic equilibrium in long-time limit, and entropy saturates to a certain value. In contrast, λ_1 always dominates the contributions for either short- or long-time regime in the case of the ratio 1:1 as shown in the lower inset of figure 1. From the statistical point of view, this somehow explains the suppressed and regular behaviour of the entanglement (entropy).

Figure 2 shows the time evolutions of the populations of the eigenstates for different ratios of pulse shapes. For 1:1 ratio, the pulse hardly excites the rotors from the initial energy level $(0, 0; 0, 0)$. Therefore, $(0, 0; 0, 0)$ is still the most populated level (the population value is nearly close to 1) as shown in the lower panel of figure 2 while the pulse passes through. Similar to the ground state, the populations of the higher levels (the inset of figure 2) also show the periodic behaviour. The periodic behaviour is ascribed to the dipole interaction. Since the small fluctuation of the population is dominated by the dipole interaction in the case of symmetrical pulses. The magnitudes of the periodic fluctuations in higher level populations are rather small with the periodic evolution of the entropy. On the other hand, for 9:1 ratio the populations of the higher states show different degrees of irregularity as shown in the upper panel of figure 2. This is because a single asymmetrical pulse can generate high populations in the excited states [6], i.e. a larger angle orientation. The larger angle orientation can cause a

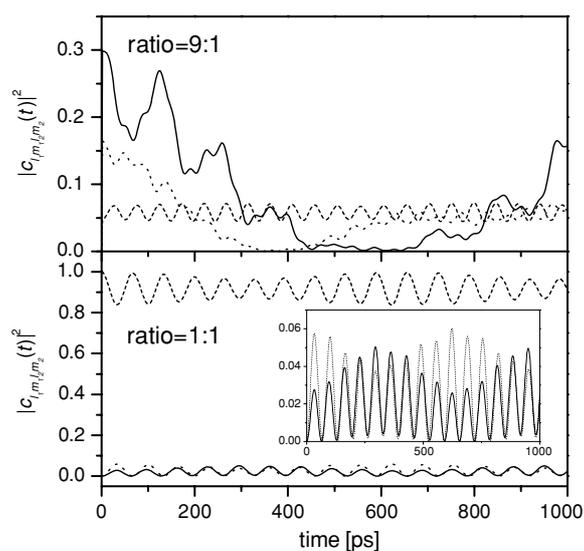


Figure 2. Populations of the states $(l_1, m_1; l_2, m_2)$ for different ratios. Upper panel: $(1, 0; 0, 0)$ (dashed line), $(1, 0; 1, 0)$ (solid line), $(2, 0; 1, 0)$ (dotted line). Lower panel: $(0, 0; 0, 0)$ (dashed line), $(1, 0; 1, 0)$ (solid line), $(1, 1; 1, 1)$ (dotted line). The inset in the lower panel is the enlarged figure showing the states $(1, 0; 1, 0)$ (solid line), $(1, 1; 1, 1)$ (dotted line), respectively.

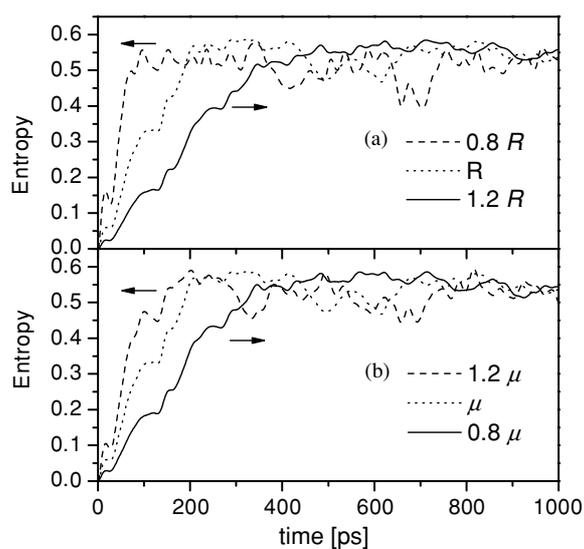


Figure 3. Time evolution of the entropy for different separation and dipole moment under single pulse (ratio 9:1). The dotted curve shows the case of $R = 1.5 \times 10^{-8}$ m and $\mu = 9.2$ D. The dashed and solid curves correspond to (a) $0.8 R$ and $1.2 R$, or (b) 1.2μ and 0.8μ , respectively.

largely fluctuated dipole interaction between the molecules. For this situation, energy transfer by means of (mediated) dipole interaction generates the irregular evolutions of the higher excited states which result in a randomly time-varying entropy.

We further study the entropy for different separation and dipole moment in figure 3. The ratio is set equal to 9:1. If the separation is smaller ($0.8 R$), the entropy grows rapidly.

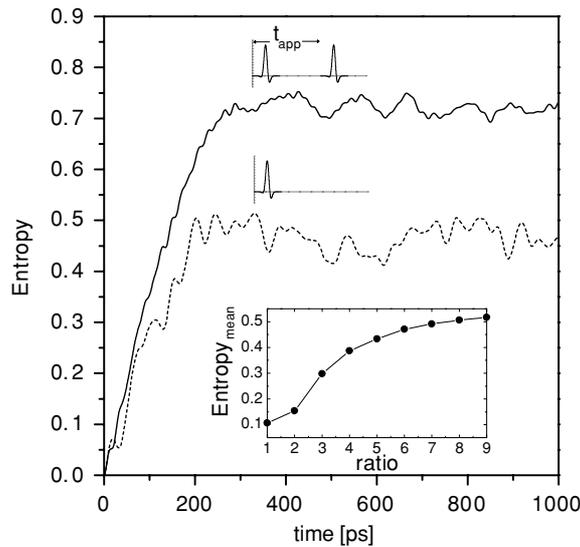


Figure 4. Time evolution of the entropy for fixed field strength $E_0 = 3 \times 10^7 \text{ V m}^{-1}$ and ratio 5:1 (frequency $9 \times 10^{11} \text{ s}^{-1}$) under single pulse (dashed line) and double pulses (solid line). Time separation (t_{app}) between two pulses is set to be five times the centre of the laser peak. Inset: dependence of the time-averaged entropy on the pulse shape for inter-molecule separation $R = 1.5 \times 10^{-8} \text{ m}$.

In contrast, the entropy evolves slowly for the case of larger separation. This means that the system needs much more time to approach the dynamic equilibrium. We also study the time evolution of entropy by changing the dipole moment. Our result shows that a similar behaviour of the entropy exhibits, i.e. the strength of dipole interaction governs the behaviour of evolution.

By adjusting the laser parameters, one can vary the degree of the entanglement. Figure 4 illustrates the time evolution of the entropy under single pulse or double pulses with ratio 5:1. As can be seen, an irregular behaviour of the entropy is obtained, but their averaged values are different. For single kick, the populations are first dominated by this laser pulse. Then, the dipole interaction plays a key role to raise the entanglement in the system. In the case of double pulses the finite populations are created by first pulse. As the second laser pulse passes through, the populations will be redistributed to a wider range. Since the populations are distributed more averagely in this case, the entropy is certainly larger as shown by the solid line in figure 4. One can note that the enhancement of entropy is achieved by applying the second laser pulse. Consider the case that the time separation between these two pulses is set to be five times the centre of the laser peak. Here we emphasize that the time separation is not fixed and can be tuned to obtain different degrees of entropy. Another way to control the degree of entanglement in this system is to change the positive and negative ratios of the laser pulse. The inset of figure 4 shows the *time-averaged* entropy with respect to different ratios. We find that the entropy is more enhanced as the ratio is larger. This means that the highly asymmetric laser pulse can generate larger entropy under the same field strength.

To study the crossover behaviour from quantum to classical limit in this system, one can tune the fundamental Planck constant \hbar' . Figure 5 shows the time for entropy first exceeds the *time-averaged* value (arrow in the inset) versus the different factors of Planck constant \hbar' . As shown, the time grows rapidly with the decrease of the Planck constant \hbar' . The inset in

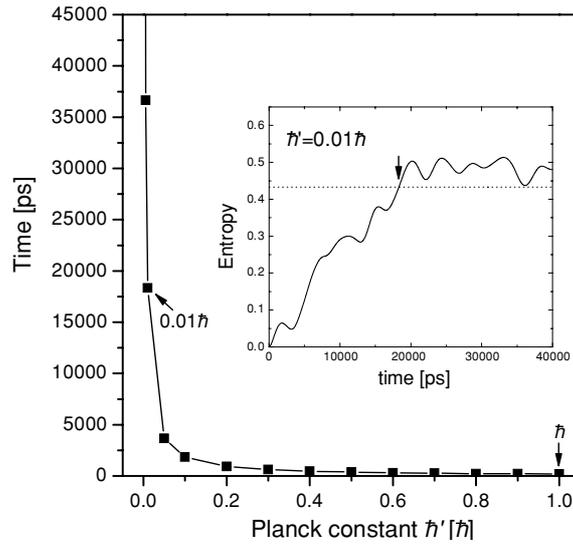


Figure 5. The time with respect to different Planck constant \hbar' for fixed field strength $E_0 = 3 \times 10^7 \text{ V m}^{-1}$, frequency $9 \times 10^{11} \text{ s}^{-1}$ and inter-molecule separation $R = 1.5 \times 10^{-8} \text{ m}$. The time is defined as the first time in entropy that exceeds the time-averaged value (the arrow in the inset). Inset: the time-averaged value (dotted line), and time evolution of the entropy for $\hbar' = 0.01\hbar$ (solid curve).

figure 5 shows a slow increase of entropy with the evolution of time for $\hbar' = 0.01\hbar$. Comparing this with the result for $\hbar' = \hbar$, the ratio of the two times is roughly 100:1. This means that the entropy evolves slowly, and the system needs a longer time to approach dynamical equilibrium for a small \hbar' . As expected, the time for classical limit ($\hbar' \rightarrow 0$) goes to infinity, satisfying that no entanglement exists between classical objects.

For a more realistic molecular system, one can extend our model to hindered-rotor system. The hindered rotor means that the polar diatomic molecule is adsorbed on the surface with the confinement of surface potential. In other words, one reasonably considers that two coupled polar molecules are adsorbed on the surface with the dipole interaction. The theoretical models about hindered rotor have been proposed by the authors [32, 35–37] and received some success in agreement with experiments [38, 39]. Comparing hindered rotor with free one, the rotation of a hindered rotor is similar to that for the free one; but the degree of orientation is different. This is because that the surface potential confines the rotation. Although this confinement may affect the property of the system, according to our previous work [40], a free rotor and a hindered rotor actually show the same physics. In particular, a hindered rotor can be transformed into a free one by changing the parameters of surface potential.

A few remarks about the experimental verifications of our model can be addressed here. The separation distance between two molecules can be modulated on the surface with the help of nanotechnologies [19–21]. Besides, previous studies have shown that the molecular axis orients in response to the distribution of populations of molecules [6]. A similar property also has been verified in the hindered-rotor model [40]. As for the measurements, it is found that the orientations of coupled rotors directly relate to the entropy according to our studies [29]. This indicates the orientations of coupled rotors somehow reflect the behaviour of the entropy of the system. Many experiments have been performed to measure the orientations of the rotors [13, 14]. Under proper arrangements, the orientations of hindered rotors can also

be measured by the similar technologies. This may provide a method to see the indication of the entanglement from orientation.

4. Conclusions

We have studied the entanglement of two coupled polar molecules irradiated by half-cycle pulses. The behaviour of the entanglement is analysed via varying the symmetry of the laser shape. Periodic-like entropy is found as a symmetrical-ratio pulse is shined, while a highly asymmetrical pulse can induce highly irregular entropy. Furthermore, the entanglement is enhanced by applying multi-pulses. We also discussed the difference between the quantum and classical regimes in this coupled-rotor system. By varying the Planck constant, crossover from quantum to classical limit can be seen from the von Neumann entropy. Prominent difference in entropy between quantum and classical regimes is the time that approaches dynamical equilibrium. In particular, for classical limit ($\hbar' \rightarrow 0$), the system is expected to need infinite time for dynamical equilibrium. These results may contribute to the essential physics of coupled rotors and may be useful in understanding the field–molecule interactions.

Acknowledgments

We would like to thank Dr T Brandes at University of Manchester for helpful discussions. This work is supported by the National Science Council, Taiwan under the grant numbers NSC 94-2120-M-009-002 and NSC 94-2112-M-009-024.

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