Suppressing the weakly bound states in the photoassociation dynamics by using a frequency cut-off laser pulse*

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The photoassociation dynamics of ultracold lithium atoms controlled by a cut-off pulse has been investigated theoretically by solving numerically the time-dependent Schrödinger equation using the mapped Fourier grid method. The frequency components of the laser pulse close to the atomic resonance are partly cut off. Compared with the typical Gauss-type pulses, the cut-off pulse is helpful to suppress efficiently the weakly bound states and prepare the associated molecules in the lower vibrational states. Especially, the dependence of photoassociation probability on the cut-off position of the laser pulse is explored.

Keywords: photoassociation, cut-off laser pulse, ultracold molecules

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1. Introduction

Photoassociation is one of the most efficient ways to produce ultracold molecules in the ultralow temperature region from millikelvin to nanokelvin.[1−11] The formation of ultracold molecules can be implemented via photoassociation in the short- or long-range potential region.[1,12,13] A two-color pump-dump photoassociation scheme has been proposed to investigate the formation of ultracold molecules with chirped pulses.[12] In most cases, photoassociated molecules are in their weakly bound states of the long-range potential.[14] Recently, the ultracold molecules in the lower or the lowest vibrational states of ground electronic state have been prepared by using the pump-dump technique.[15,16]

With the development of laser techniques,[17,18] laser pulses now can be well shaped.[3,19,20] The shaping technique has been widely applied to the coherent control of an ultracold molecular system.[21−24] In the photoassociation with ultrashort laser pulses, the pulses have a very broad frequency width, which can cause both molecular and atomic transitions simultaneously. It is necessary to cut off the spectrum of the laser pulse at the atomic resonance frequency to suppress the atomic transition. Cutting the laser pulse can lead to a long tail in the time domain (usually in the picosecond scale)[25,26] and induce the coherent transient dynamics.[27,28] This kind of pulse has been used to steer the photoassociation of ultracold rubidium atoms.[26] However, most of the associated molecules still reside at the weakly bound states. Hence, the scheme used for controlling photoassociation needs to be improved.

In the present work, we investigate theoretically the photoassociation dynamics of ultracold lithium atoms steered by a cut-off pulse using the wavepacket dynamics method, aiming at suppressing the weakly bound states.

2. Model and method

At temperature T, two cold lithium atoms collide in the ground state potential and form the Li_2 molecule in the electrical excited state, as shown in Fig. 1. The time-dependent Schrödinger equation can be expressed as

$$\mathrm{i}\hbar\frac{\partial}{\partial t}\begin{pmatrix}\Psi_{\mathrm{g}}(R,t)\\ \Psi_{\mathrm{e}}(R,t)\end{pmatrix}=\begin{pmatrix}\hat{T}+V_{\mathrm{g}}(R) & \hat{W}(t)\\ \hat{W}(t) & \hat{T}+V_{\mathrm{e}}(R)\end{pmatrix}$$

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$$\times \begin{pmatrix} \Psi_{\rm g}(R,t) \\ \Psi_{\rm e}(R,t) \end{pmatrix}, \qquad (1)$$

where $\Psi_{\rm g}(R,t)$ and $\Psi_{\rm e}(R,t)$ denote the wave functions of $1^3\Sigma_{\rm u}^+$ and $2^3\Sigma_{\rm g}^+$ states, respectively, $\hat{T}$ is the kinetic energy operator, $V_{\rm g}(R)$ and $V_{\rm e}(R)$ denote the potential functions of $1^3\Sigma_{\rm u}^+$ and $2^3\Sigma_{\rm g}^+$ states, respectively.

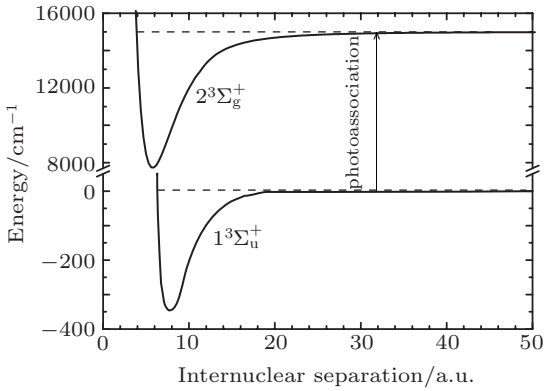


Fig. 1. Potential curves of the ground state $1^3\Sigma_{\rm u}^+$ and the excited state $2^3\Sigma_{\rm g}^+$ for the lithium dimer and the photoassociation process. The dashed line directly above the $1^3\Sigma_{\rm u}^+$ state denotes the initial continuum level.

An accurate calculation is performed for Eq. (1) without employing the rotating-wave approximation. The interaction $\hat{W}(t)$ is written in the dipole approximation as

$$\hat{W}(t) = -\hat{\varepsilon}(t)\cdot\hat{\mu}(R) = -W_0 f(t), \qquad (2)$$

where $W_0 = \varepsilon_0\mu(R)$, and $\mu(R)$ is the R-dependent dipole moment operator. The rescaling time-dependent electric field of the cut-off pulse, $f(t)$, is given by

$$f(t) = C_0\,{\rm Re}\left[\int_{-\infty}^{\infty} A(\omega)\,{\rm e}^{{\rm i}\phi(\omega)}\,{\rm e}^{-{\rm i}\omega t}\,{\rm d}\omega\right], \qquad (3)$$

where

$$A(\omega) = \begin{cases} \exp[-2\ln 2(\omega-\omega_0)^2/\tau_{\rm f}^2], & \omega < \omega_{\rm cut}, \\ 0, & \omega \geq \omega_{\rm cut}, \end{cases} \qquad (4)$$

is the real-valued Gaussian distribution of frequencies with full width at half maximum (FWHM) $\tau_{\rm f}$. In Eqs. (3) and (4), C_0 is a scaling factor, $\omega_{\rm cut}$ is the cut-off position of the laser pulse in the frequency domain, and $\phi(\omega)$ denotes the phase of the laser pulse. In the present work, $\phi(\omega)$ is chosen to be independent of frequency ω, i.e., $\phi(\omega)$ is taken to be zero for simplicity. The spectral components of the laser pulse close to or beyond the dissociation threshold of the excited state potential are cut off in order to suppress the atomic transition. The spectral intensities of three kinds of laser pulses (cut-off pulse, fat and slender Gauss-type pulses) with the same energy versus frequency are shown in Fig. 2(a), and the corresponding temporal envelopes are depicted in Fig. 2(b).

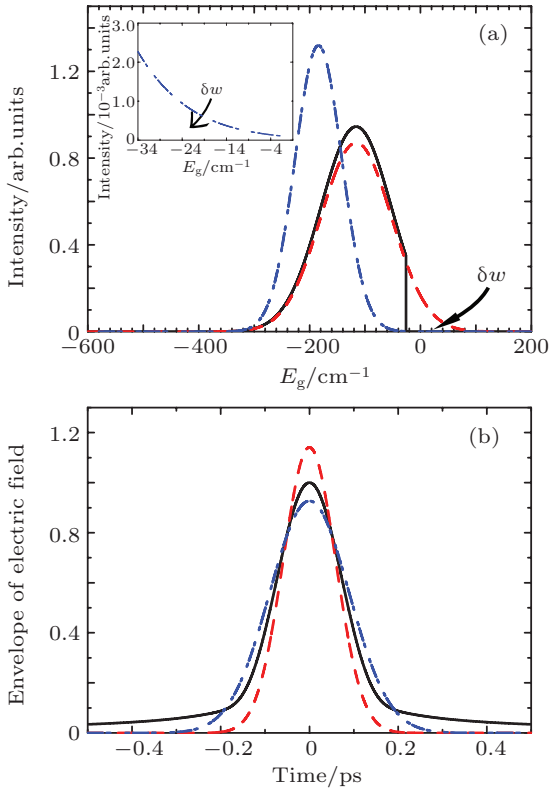


Fig. 2. (colour online) (a) Spectral intensities of the fat Gauss-type pulse (dash line), the cut-off pulse (black solid line), and the slender Gauss-type pulse (dot dash line). The inset shows the intensities in the vicinity of $E_{\rm g} = -26$ cm^{-1}. (b) Temporal envelopes of the three kinds of laser pulses in panel (a).

In our calculation, the dipole moment of the Li–Li complex is adopted from Ref. [29]. The short-range potential curves are taken from Ref. [30], which are matched to the long-range dispersion potentials with C_n coefficients.[31,32] By diagonalizing the Hamiltonian in Eq. (1) on a mapped Fourier grid,[33,34] we can obtain the accurate vibrational wave functions. The photoassociation dynamics is described by numerically solving the time-dependent Schrödinger equation using the Chebyshev propagator.[35]

3. Results and discussion

The numerical calculations are performed with the size of $L_R = 15000a_0$ at the temperature of $T = 53$ μK, corresponding to the energy of 1.68×10^{-10} a.u.

(3.69×10^{-5} cm^{-1}). The propagation time of wave packet at the excited electronic state is taken to be 20 ps, which is much shorter than the lifetime of the atomic (Li) level and the radiative lifetime of the vibrational levels of the $2^3\Sigma_g^+$ state. Therefore, we can neglect the loss caused by the spontaneous emission here. In our calculation, the temperature is so low that we only need to consider the contribution of the s-wave to the photoassociation dynamics.

The Franck–Condon factors between the bound vibrational states of the excited $2^3\Sigma_g^+$ electronic potential and the initial scattering state of the ground state potential are calculated as a function of the vibrational quantum number v, as shown in Fig. 3. The Franck–Condon factors vary over several orders of magnitude and have an obvious influence on the final population distribution. The highest peak emerges at around $v = 105$, which is also the location of the maximal population under the action of Gaussian pulses.

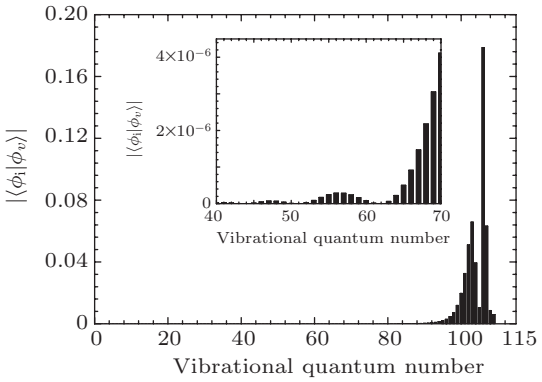


Fig. 3. The Franck–Condon factors between the bound vibrational states ϕ_v of the excited $2^3\Sigma_g^+$ electronic potential and the initial scattering state ϕ_i of the ground state potential. The inset shows the results for v from 40 to 70.

First, we utilize a fat Gauss-type pulse to steer the photoassociation process. This kind of laser pulse has its central frequency at $E_g = -116$ cm^{-1} and FWHM $\tau_f = 150$ cm^{-1} in the frequency domain. Here E_g is defined as $E_g = E_e^n - V_e(\infty) = -E_b$, where E_e^n denotes the eigenenergy of the n-th vibrational state, $V_e(\infty)$ is the energy at the atomic resonance frequency, and E_b is the binding energy. When the pulse has passed through, the population distribution is shown in Fig. 4(a). We can see that a strong peak appears at $v = 105$, and there is hardly any population at the lower vibrational levels. This is not what we pursue, because the higher excited molecules are extremely unstable.

Our goal is to control the photoassociation population distribution in the lower vibrational bound states rather than that in the weakly bound states close to the dissociation limit. Thus, the components of the pulse frequency close to the dissociation limit should be cut off. The center frequency of the cut-off pulse is taken to be the same as that of the fat Gauss-type pulse, and its peak intensity is chosen to be 1.0×10^7 V/m. The spectrum of the laser pulse is cut off at $E_g = -26$ cm^{-1}. After the pulse has passed through, the vibrational population distribution is shown in Fig. 4(b). We can observe from Fig. 4(b) that the maximum of the population appears at $v = 69$, and most of the population accumulate in the region of v from 53 to 69 rather than that from 97 to 106 shown in Fig. 4(a). The population distribution in the weakly bound states is also shown in Fig. 4(b), from which we can see that the maximal population is 2.63×10^{-10}. Moreover, with the cut-off pulse, the total population in the lower levels rises to 3.84×10^{-8}, which is nearly 1.2 times of that with the fat Gauss-type pulse. Especially, the total population in the weakly bound states decreases by six orders of magnitude, from 4.42×10^{-4} to 7.21×10^{-10}. Therefore, we can reduce the trouble caused by the intensely oscillating Franck–Condon factors and prepare stable associated molecules with the cut-off pulse. Compared with the fat Gauss-type pulse, the cut-off pulse can efficiently suppress the weakly bound states.

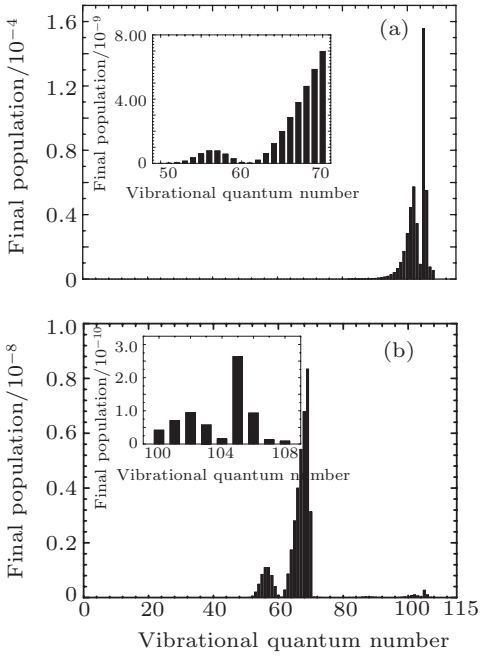


Fig. 4. Population distributions in the excited $2^3\Sigma_g^+$ states steered by (a) the fat Gauss-type pulse and (b) the cut-off pulse. The insets in panels (a) and (b) show the results for v from 50 to 70 and from 100 to 108, respectively.

To fully compare the validity of the cut-off pulse in suppressing the weakly bound states with that of the typical Gauss-type pulses, we also utilize a slender Gauss-type pulse to implement the photoassociation process. As shown in Fig. 2(a), the slender Gauss-type pulse possesses a similar frequency distribution to that of the cut-off pulse, but its intensity in the vicinity of $E_g = -26$ cm^{-1} is very low (see the inset in Fig. 2(a)). The central frequency is at $E_g = -184$ cm^{-1}, and the FWHM is $\tau_f = 99$ cm^{-1} in the frequency domain. After the pulse has passed through, the result is illustrated in Fig. 5, from which we find that most of the population still stay at the vibrational levels from v=100 to 106. In short, with the typical Gauss pulses, even if its intensity is extremely small, we still can not suppress the weakly bound states and prepare stable associated molecules.

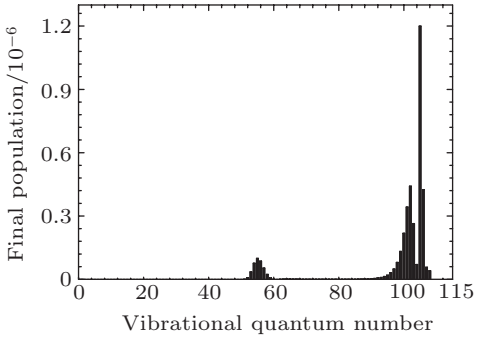


Fig. 5. Population distribution in the excited $2^3\Sigma_g^+$ states steered by the slender Gauss-type pulse.

We are interested to know the dependence of photoassociation probability on the cut-off position of the laser pulse. The cut-off position ΔE denotes the shift from the cut-off line to the central frequency. In our calculation, we allow ΔE varying in the range from 0 cm^{-1} to 105 cm^{-1}. Figure 6 illustrates the total population versus ΔE. When the laser pulse is cut off in the the range from $\Delta E = 0$ cm^{-1} to 45 cm^{-1}, a small population is obtained, leading to a lower photoassociation probability. However, when the cut-off position is in the range from 75 cm^{-1} to 95 cm^{-1}, a larger population is obtained. In the inset of Fig. 6, we also plot the population difference Δp between the lower vibrational states and the weakly bound states. The best cut-off position of the laser pulse should be in the vicinity of the maximal value of Δp. Based on the above calculated results, We think that the cut-off position of the laser pulse should lie in the range of from 90 cm^{-1} to 100 cm^{-1} to suppress the weakly bound states and prepare the stable associated molecules efficiently.

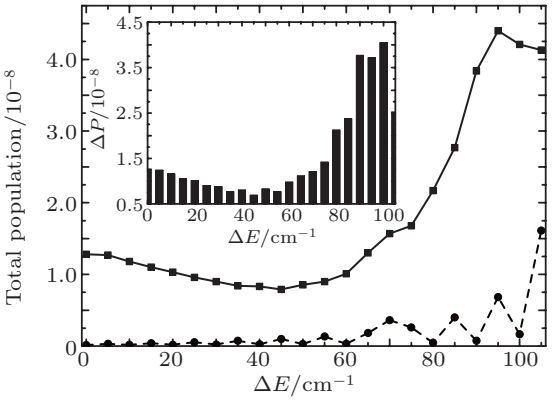


Fig. 6. Total populations of the lower vibrational states (from $v = 50$ to 70, marked by solid line) and the weakly bound states (from $v = 90$ to 108, marked by dashed line) versus ΔE. The inset shows the population difference Δp between the lower vibrational states and the weakly bound states.

4. Conclusion

We have studied theoretically the formation of ultracold Li_2 molecules via photoassociation with a cut-off pulse. For comparison, we also utilize the typical Gauss-type pulses to steer the photoassociation process. With the cut-off pulse, the weakly bound states can be suppressed, and the stable associated molecules in the lower vibrational levels can be produced. With the typical Gauss-type pulse, whether it is the fat or slender type, most of the population stay at the higher vibrational levels close to the dissociation threshold. In addition, we also analyze the dependence of photoassociation probability on the cut-off position of the laser pulse.

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