

Light-induced conical intersection effect enhancing the localization of molecules in optical latticesMariusz Pawlak^{1,2} and Nimrod Moiseyev^{1,3}¹*Schulich Faculty of Chemistry, Technion-Israel Institute of Technology, Haifa 32000, Israel*²*Faculty of Chemistry, Nicolaus Copernicus University in Toruń, Gagarina 7, 87-100 Toruń, Poland*³*Faculty of Physics, Technion-Israel Institute of Technology, Haifa 32000, Israel*

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The cold weakly bound Rb₂ molecule dressed by a standing linear polarized laser wave is investigated. In such a case the light-induced conical intersections (LICIs) occur. We demonstrate that when the LICI conditions are fulfilled the transition from the weakly trapped rubidium dimer in the red-detuned optical lattice to the strongly trapped rubidium dimer in the blue-detuned optical lattice takes place. A significant localization enhancement of the molecule is observed.

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I. INTRODUCTION

The conical intersection (CI) problem has been known for many decades. It was first recognized by von Neumann and Wigner [1,2]. The discovery of this phenomenon in photochemistry is attributed to Zimmerman [3] and Michl [4]. Nowadays, we know how important a role CIs play in chemical reactions and different molecular processes. For detailed reviews see, for example, Refs. [5–8] and references therein.

CIs between two (or more) potential-energy surfaces are created due to the couplings between nuclei and electrons of a system. They provide funnels for ultrafast radiationless interstate transitions. In the vicinity of CIs the nonadiabatic couplings are large, and the Born-Oppenheimer approximation does not hold. Interesting and unanticipated new effects occur. It means that some experimental results can be explained only when the Born-Oppenheimer approximation breaks down for meeting points of two potential surfaces. Due to the strong couplings between electrons and nuclei the dynamics can be considered based on the quantum theory only. The classical theory does not give the correct description of nonadiabatic dynamics.

The importance of CIs in the theory of chemical reactions is fundamental. Moreover, many effects are related to the existence of CIs, e.g., an isomerization, a relaxation dynamics, and a proton transfer [9]. CIs are found in various areas of chemistry, physics, biology, and nanosciences. For example, they are well known in organic chemistry [10–14], spectroscopy [9], solid-state physics [15], and optics [16,17]. The rapid development of experimental femto- and attosecond laser techniques enables accurate measurements and provides evidence for CIs in new circumstances.

CIs can be observed in polyatomic molecules consisting of at least three nuclei, i.e., the system requires at least two independent nuclear degrees of freedom whose variation modifies the electronic function. For a diatomic molecule if two electronic states belong to the same irreducible representations then the corresponding potential-energy curves cannot cross. In other words, two states cannot be degenerate when the corresponding wave functions have the same symmetry. Such a molecule has only one degree of freedom (distance between nuclei—nuclear vibrational coordinate) which is not enough to provide a branching space. The situation looks different

when the system is under the influence of an external field, e.g., a magnetic field or an electric field. If the laser field is applied the CIs can be formed in all molecules even in the diatomic ones, and the additional degree of freedom appears. As a result of the interaction of the dipole moment of the system with the laser field an effective torque towards the field polarization direction is generated [18]. Thus, the diatomic molecule may rotate. For a running-wave laser field the laser induces the CIs which couple the rotational and vibrational motions. In turn, for a standing-wave laser field the laser induces the CIs which also couple the motion of the center of mass with all internal (electronic, vibrational, and rotational) motions. These intersections are called light- or laser-induced conical intersections (LICIs). They can be controlled by manipulating the laser field frequency and intensity. The LICI effect was discovered theoretically several years ago [19]. So far, there has been increasing interest in investigations of the phenomenon of the LICI primarily in diatoms (where Na₂ or D₂⁺ was taken as a representative system) [19–28] but also in polyatomic molecules [29,30].

In this paper, in contrast to previous studies, we consider a two-atom homonuclear cold weakly bound rubidium molecule. Recently, rubidium atoms were intensively used in research on highly excited states [31–33], Rydberg atom blockade [34,35], creating diatomic molecules [36–38], etc. The investigated system is subjected to the influence of a standing linearly polarized laser field forming a one-dimensional optical lattice. Our main goal is to find out what the effect of the light-induced conical intersection on the molecular localization is. The theoretical simulations involve the ground and excited electronic states and are based on the Floquet approach.

II. THEORETICAL BACKGROUND

Let us describe the form of the Hamiltonian. We represent the time-dependent Hamiltonian in the Floquet picture [39–41] using formalism as in Ref. [19]. The standing laser wave is formed by interfering two running waves [42]: $\varepsilon_0[\sin(k_L Z + \omega_L t) + \sin(k_L Z - \omega_L t)]/2 = \varepsilon_0 \sin(k_L Z) \cos(\omega_L t)$, where ε_0 is the laser field amplitude and k_L is the wave vector associated with the laser frequency ω_L by relation $k_L = \omega_L/c$. When we assume that only the single photon is absorbed by the

system, the Hamiltonian reduces to the following static (time-independent) matrix form:

$$\mathbf{H} = \mathbf{T} + \mathbf{V} = \begin{pmatrix} \hat{T}_{X,Y,Z} + \hat{T}_{R,\theta,\phi} & 0 \\ 0 & \hat{T}_{X,Y,Z} + \hat{T}_{R,\theta,\phi} \end{pmatrix} + \begin{pmatrix} V_g(R) & \frac{\varepsilon_0}{2} d(R) \cos \theta \sin(k_L Z) \\ \frac{\varepsilon_0}{2} d(R) \cos \theta \sin(k_L Z) & V_e(R) - \hbar\omega_L \end{pmatrix}. \quad (1)$$

Such a constructed single-channel Hamiltonian enables explaining in a simple way many effects in chemistry and physics in laser fields [19,20,22–24,29,30,38,42]. The first matrix in Eq. (1) contains the kinetic-energy operators for the translational motion (of the center of mass) $\hat{T}_{X,Y,Z}$ and the rotational-vibrational motion $\hat{T}_{R,\theta,\phi}$. These terms are as follows:

$$\hat{T}_{X,Y,Z} = -\frac{\hbar^2}{2M} \left(\frac{\partial^2}{\partial X^2} + \frac{\partial^2}{\partial Y^2} + \frac{\partial^2}{\partial Z^2} \right), \quad (2)$$

$$\hat{T}_{R,\theta,\phi} = -\frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial R^2} + \frac{\hat{L}^2(\theta,\phi)}{2\mu R^2}, \quad (3)$$

where M is the total mass of the system and μ is the reduced mass of the two atoms. The next matrix in Eq. (1) represents the potential part of the Hamiltonian where $V_g(R)$ and $V_e(R)$ are field-free Born-Oppenheimer potentials of the ground $(1)^1\Sigma_g^+$ and excited $(1)^1\Sigma_u^+$ electronic states, respectively, which are coupled by the applied electromagnetic field. The diabatic potential-energy curves (PECs) are presented in Fig. 1. The light is propagated along the Z direction. The laser frequency is held to be red detuned (RD) from the atomic resonance $\omega_L = 0.057\,681\,5$ a.u. ($\lambda = 790$ nm). The transition dipole moment $d(R)$ as well as the potentials $V_g(R)$ and $V_e(R)$ are taken from Ref. [43]. The dressing of electronic states

by the laser field is qualitatively different from the atomic case due to the existence of the dynamical variables θ and R . The angle θ (one of the rotational coordinates) is the orientation of the diatomic molecule with respect to the light propagation axis; R is the distance between atoms. We assume that the initial molecular state that is chosen as the electronic and rotational ground state is coupled with electronically excited state and $J = 1$. Therefore, the initial orientation of the diatom is fixed during the process. Hence, the effective field amplitude is $\varepsilon_0^{\text{eff}} = \varepsilon_0 \langle \psi_{J=0}^g | \cos \theta | \psi_{J=1}^e \rangle$. In other words, this condition implies that the molecular rotation is kept frozen in the experiment. This is a well-known approach that is often taken into consideration [26–28,44]. We assume the mass of a single Rb atom $m_{\text{Rb}} = 85.467\,8$ u (in a.u.: 155 798.268) as it is given by Allouche and Aubert-Frécon [43]. The laser field intensity is $I_0 = 3.51 \times 10^8$ W/cm² ($\varepsilon_0 = 10^{-4}$ a.u.). There has been increasing interest in theoretical investigations of diatoms under the influence of running laser waves with such an intensity [20,22–25]. Note that experiments were carried out successfully for two-atom systems in optical lattices for lower as well as for greater values of laser field intensity, see, e.g., Refs. [45,46], respectively.

The diabatic Hamiltonian (1) can be transformed to adiabatic form where the kinetic part contains the coupling elements, whereas the potential part is diagonal. Thus, by carrying out a quantum-mechanical unitary transformation $\begin{pmatrix} \cos \Phi & -\sin \Phi \\ \sin \Phi & \cos \Phi \end{pmatrix}^\dagger \begin{pmatrix} V_{11} & V_{12} \\ V_{21} & V_{22} \end{pmatrix} \begin{pmatrix} \cos \Phi & -\sin \Phi \\ \sin \Phi & \cos \Phi \end{pmatrix}$, where $V_{21} = V_{12}$ and $\tan(2\Phi) = 2V_{12}/(V_{11} - V_{22})$, the two adiabatic potential-energy surfaces $W_{\text{upper}}(Z, R, \theta)$ and $W_{\text{lower}}(Z, R, \theta)$ occur. They may meet causing the LICIs provided that the following conditions are simultaneously fulfilled: (a) $V_g(R) = V_e(R) - \hbar\omega_L$; (b) $Z = n\pi/k_L$ (where n is an integer) or $\theta = \pi/2$. It means that the off-diagonal elements in the potential matrix \mathbf{V} are equal to zero, and the diagonal ones are the same. Figure 2 illustrates adiabatic potential-energy surfaces against Z/λ

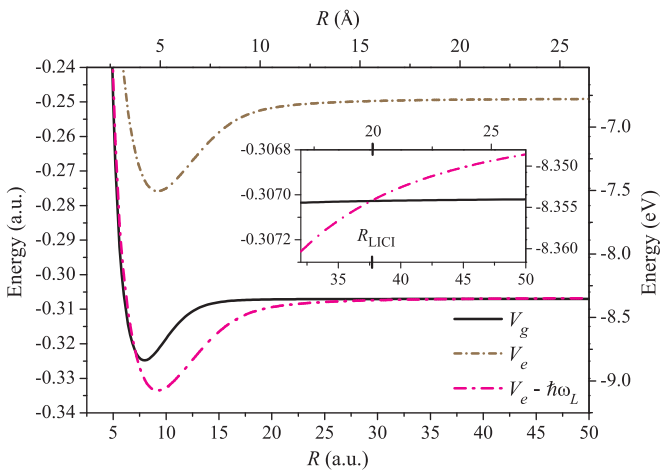


FIG. 1. (Color online) Field-free potential-energy curves $V_g(R)$ and $V_e(R)$ associated with the $(1)^1\Sigma_g^+$ and $(1)^1\Sigma_u^+$ electronic states, respectively, of Rb_2 . The field-free PEC associated with the $(1)^1\Sigma_u^+$ state shifted by the single-photon energy is also presented. The laser frequency is held at $\omega_L = 0.057\,681\,5$ a.u.

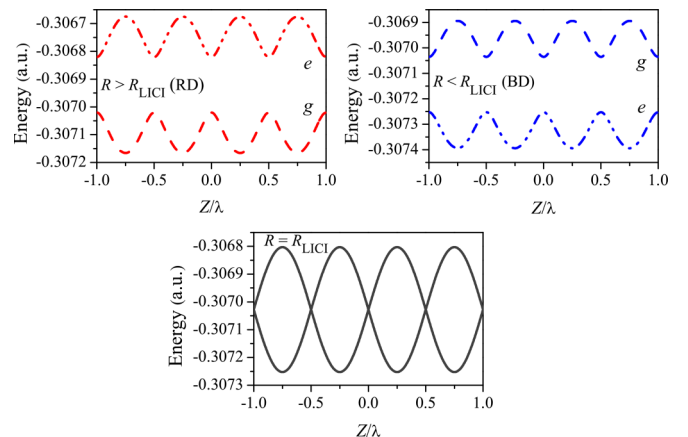


FIG. 2. (Color online) Adiabatic potential-energy surfaces $W_{\text{upper}}(Z, R, \theta = 0)$ and $W_{\text{lower}}(Z, R, \theta = 0)$ for $R = 50$ a.u. (upper-left panel) for $R = R_{\text{LIC1}} = 37.6195$ a.u. (lower panel) and for $R = 32$ a.u. (upper-right panel) of the weakly bound Rb_2 molecule dressed by a standing laser wave. Each of the surfaces is composed of two parts corresponding initially to diabatic states: ground (g) and excited (e). The laser intensity and frequency are held at $I_0 = 3.51 \times 10^8$ W/cm² and $\omega_L = 0.057\,681\,5$ a.u., respectively.

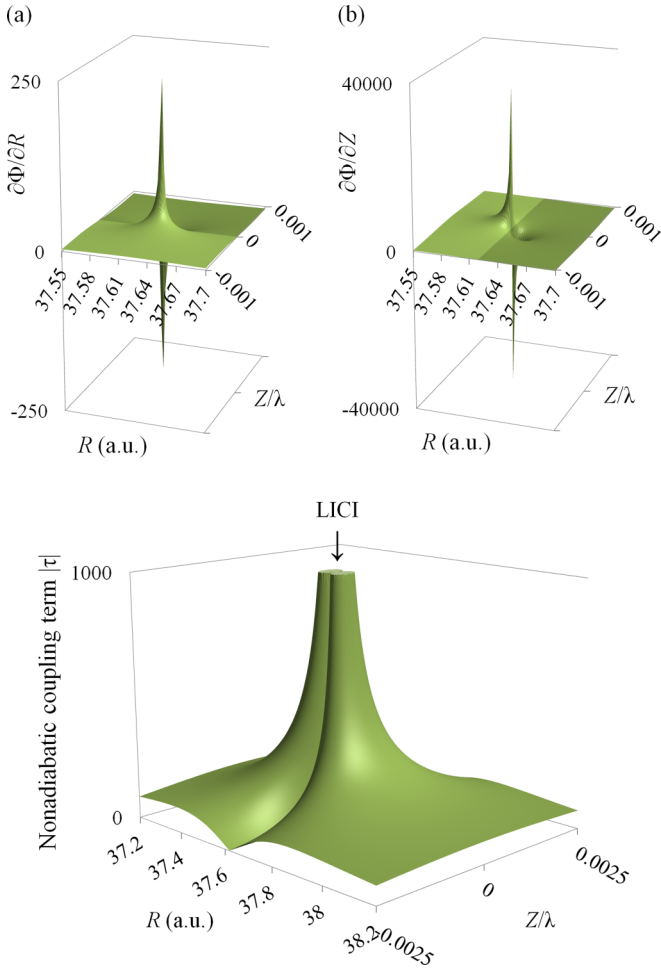


FIG. 3. (Color online) Nonadiabatic coupling terms: (a) $\tau_R = \partial\Phi(Z, R, \theta = 0)/\partial R$, (b) $\tau_Z = \partial\Phi(Z, R, \theta = 0)/\partial Z$, and $|\tau| = \sqrt{\tau_R^2 + \tau_Z^2}$ (lower panel) for the weakly bound Rb_2 in the vicinity of the LICI. The laser intensity and frequency are held at $I_0 = 3.51 \times 10^8 \text{ W/cm}^2$ and $\omega_L = 0.0576815 \text{ a.u.}$, respectively.

for the intermolecular distance $R = 50 \text{ a.u.}$ (26.4589 \AA), $R = R_{\text{LICI}}$ which in our case is 37.6195 a.u. (19.9074 \AA), and $R = 32 \text{ a.u.}$ (16.9337 \AA). The latter case corresponds to a blue-detuned (BD) standing laser wave. The surfaces exhibit a periodic chain of intersecting points for $R = R_{\text{LICI}}$ and $Z/\lambda = \dots, -1, -0.5, 0, 0.5, 1, \dots$

After our transformation of the Hamiltonian (1) to adiabatic representation the nonadiabatic couplings appear in the kinetic energy. These couplings can be calculated as the derivatives of the mixing angle $\Phi(Z, R, \theta)$ in $\tan(2\Phi) = \frac{\epsilon_0 d(R) \cos \theta \sin(k_L Z)}{V_g(R) - V_e(R) + \hbar\omega_L}$ with respect to the corresponding nuclear coordinate [19]. We calculated the nonadiabatic coupling terms $\tau_R = \partial\Phi(Z, R, \theta = 0)/\partial R$ and $\tau_Z = \partial\Phi(Z, R, \theta = 0)/\partial Z$ for the rubidium dimer by numerical integration using the five-point finite difference method. Our results presented in Fig. 3 show strong nonadiabatic couplings around the LICI. The motion of the center of mass of the system is not separable from its internal motions. It is clear that the value of $|\tau| = \sqrt{\tau_R^2 + \tau_Z^2}$ becomes infinite at the exact LICI point. This situation corresponds to conical intersections that appear in field-free

molecules consisting of more than two atoms. When two potential-energy surfaces reach each other the nonadiabatic couplings become singular, the Born-Oppenheimer approximation is meaningless, and the unanticipated quantum effects can be observed.

III. RESULTS AND DISCUSSION

We are interested in the impact of the LICIs on the trapping process of cold weakly bound Rb_2 molecules. For the sake of simplicity, we take in our studies the R -adiabatic approach, i.e., we solve the Schrödinger equation with the Hamiltonian (1) where R is treated as a parameter (and not as a dynamical variable). We neglect the nonadiabatic couplings between internal motions, but we still hold much stronger nonadiabatic couplings associated with the translational motion of the center of mass [see Figs. 3(a) and 3(b)]. We performed calculations by applying the particle in a box technique with the Fourier basis. For each potential surface 2001 functions were used.

First, we computed variationally the eigenspectrum for long distances between atoms. The available theoretical data (obtained based on *ab initio* calculations) of $V_g(R)$, $V_e(R)$, and $d(R)$ exist up to $R = 50 \text{ a.u.}$ [43]. Thus, initially our system is weakly bound with the long bond length ($R = 50 \text{ a.u.}$). The contribution of the field-free ground electronic state to all eigenstates associated with the lower adiabatic potential-energy surface (W_{lower}) is very large, more than 99.9%. Our results show that the ground electronic state is the most populated in the bound molecular eigenstate whose energy level lies just above the top of the optical potential barrier, see Fig. 4. The probability of finding the molecule at a given position is presented in Fig. 5(a). The weak localization of the center of mass is clearly seen. Due to the weak van der Waals force the atoms slowly approach one another. We carried out a set of calculations decreasing R and followed the eigenstate with the largest contribution of the ground electronic state. Note that when we get closer to the LICI point, the shape of

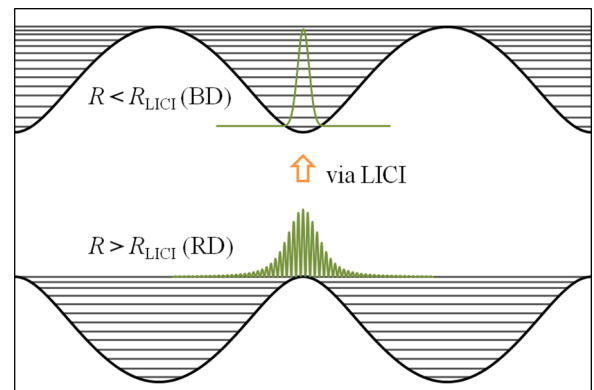


FIG. 4. (Color online) Schematic showing the effect of the LICI on the localization of the Rb_2 van der Waals molecule dressed by a standing laser wave. Initially the system where atoms are spaced apart ($R > R_{\text{LICI}}$) is in a vibrational state close to the threshold. Due to a weak attractive force the atoms approach each other. The transition from the RD to the BD optical lattice via the LICI affects the localization enhancement.

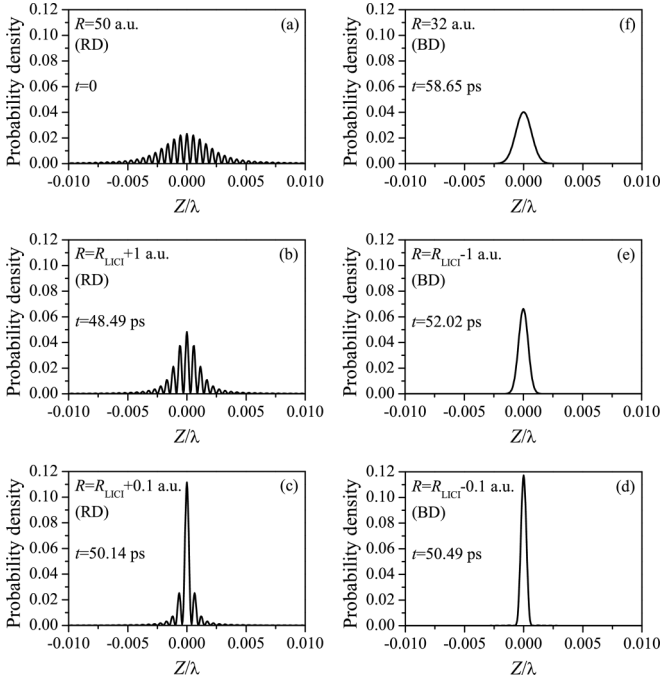


FIG. 5. Probability of finding the cold weakly bound molecule at position Z for the eigenstate with the largest contribution of the ground electronic state for different values of R , where $R_{\text{LICI}} = 37.6195$ a.u. The time of evolution due to the weak van der Waals force is also presented in each panel.

the optical lattice changes and surfaces W_{lower} and W_{upper} come up to each other. The wave function becomes narrower, and thus the system becomes more bound [see Figs. 5(a)–5(c)]. When the LICI conditions are fulfilled the transition from the weakly trapped rubidium dimer in the RD optical lattice to the strongly trapped system in the BD optical lattice takes place. The significant localization of the van der Waals molecule is observed. Now, the ground electronic state is the most populated in the lowest eigenstate associated with the upper adiabatic potential-energy surface (W_{upper}). Figures 5(a)–5(f) show strong evidence for the trapping effect via the LICI. It is clear that the light-induced strong nonadiabatic couplings dominate the molecular localization. Figure 4 presents the schematic of the enhancement of localization of cold rubidium dimer inside the optical lattice by the LICI effect. It should be noted that this phenomenon may appear only for molecules (not in the case of atoms).

In order to introduce a dynamics to our investigations, we calculated using a simple theoretical approach the time after which the population transfer through the LICI occurs starting from $R = 50$ a.u. ($t = 0$). Based on Newton’s second law of motion and the fact that the gradient of a potential gives the force acting between the atoms [$a = (-1/\mu)(dV_g(R)/dR)$, $R(t) = R_0 + v_0 t + at^2/2$], we performed numerical calculations of time applying the five-point finite difference method with dense grid spacing $\Delta R = 0.0001$ a.u. It turned out that the effect happens after about 50 ps. Such a long time is due to the weak van der Waals interaction between cold atoms that initially are very far away from each other. The appropriate time labels for

six values of R are embedded in Figs. 5(a)–5(f). Note that the spontaneous emission does not play a role because the Rb 5p state lifetime is 27 ns [47].

Furthermore, we estimated the time for a more typical experimental case where an interparticle spacing in a cold rubidium gas is much larger than 50 a.u. by fitting the Lennard-Jones potential to the available data for $R \geq R_{\text{LICI}}$. We obtained that for the distance between atoms $R = 1000$ a.u. (529.177 Å) the time it takes to get R_{LICI} is about 9.5 μs . However, the detailed analysis is beyond the scope of this paper.

Let us briefly comment on the possible realization of an experiment. First, we need to prepare by interfering counterpropagating laser beams the red-detuned one-dimensional optical lattice where rubidium atoms are trapped individually (one atom per site). Then, by shaking the lattice or modulating the standing wave (see, for example, Refs. [48–51]), we create the van der Waals dimer with the bond length of $R = 50$ a.u. and excite (nonelectronically but vibrationally) the center-of-mass motion of the molecule in the optical lattice to prepare a wave packet centered at $Z/\lambda = 0$. The van der Waals attractive force makes the two atoms move to each other.

IV. CONCLUSION

To summarize, we have theoretically investigated the cold weakly bound rubidium dimer inside the one-dimensional optical lattice. The dressing of the system by a standing laser wave causes the light-induced conical intersections, which constitute an interesting light-matter phenomenon. The presence of such LICIs is responsible for the strong nonadiabatic couplings between the translational motion of the center of mass of the diatom and all internal motions of the diatom. We have found out that the transition from the red-detuned to the blue-detuned optical lattice through the LICI significantly affects the localization enhancement of the Rb_2 molecule. Consequently, the system becomes more stable. It should be pointed out that our finding can be easily extended to other diatomic molecules and should be observed for different values of field intensity. The positions of the LICIs are determined by the frequency of the laser. Definitely, this phenomenon can be important in quantum controlling and trapping molecules in new areas of cold chemistry and physics in laser fields. We believe that our results will spur experimental studies and inspire advanced theoretical research including fine- and hyperfine-structure effects.

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