

Intense laser-controlled quenching of molecular fragmentation

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We show how efficient control can be exerted on molecular dissociation by adiabatic shaping of an intense, single laser pulse in a very simplified way, involving a few parameters (e.g., peak amplitude, carrier-wave, and modulation phases and frequencies). The strategy takes advantage of an appropriate combination of basic multiphoton mechanisms: bond softening (BS), vibrational trapping (VT), and dynamical dissociation quenching (DDQ). The resulting stabilization enhancement with respect to dissociation may increase up to a factor of 5, while simultaneously inhibiting the BS mechanism and reenforcing the VT mechanism, using DDQ. The photodissociation of H_2^+ submitted to a uv-visible field modulated by an IR periodic envelope, on which the consequences of a DDQ-type mechanism has recently experimentally been checked, is taken as an illustrative example.

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I. INTRODUCTION

By applying forces that, in energetic terms, are comparable to electron binding energies, intense laser fields produce strong internal distortions in molecules and unexpected dynamical effects that can be exploited in designing control strategies. Thus while it appears quite natural that the strong radiative interactions would generally facilitate molecular fragmentation, through the bond softening (BS) process in particular, the dissociation may, under specific conditions, be delayed or even suppressed through the complementary, non-intuitive, vibrational trapping (VT) [1,2] or, in a different frequency range, the dynamical dissociation quenching (DDQ) [3,4] mechanisms. Among them, BS and VT have been well-documented [5–7], whereas a first experimental confirmation has only been made in recent years for DDQ [8]. Two recent intense-field pump-probe experiments are also related to this effect [9]. A clear delineation of the role of the aforementioned basic mechanisms and an understanding of the way they act either in a complementary or in an antagonistic manner, are of utmost importance for the control of molecular dynamics and reactivity: With an appropriately shaped laser pulse, one may wish to soften some chemical bonds while, simultaneously, hardening others. Can one bring together the two classes of basic mechanisms (BS and VT on one hand, DDQ on the other hand) operating at different wavelength regimes with a single laser pulse, in order to enhance a desired outcome? Considering the enhancement of the stabilization with respect to dissociation as an objective, we illustrate in this paper how the interplay of these processes may be exploited, opening the route to a generic control pathway of molecular dissociation which uses pulse shaping in a very simple manner by the determination of but a few parameters (intensity, phase, carrier-wave, and modulation frequencies).

In the spectral region ranging from visible to the far ultraviolet (uv), the laser carrier-frequency is high as compared to molecular vibrational frequencies. As a consequence, the molecule feels a cycle-averaged force field. This leads to the light induced potentials of the time-independent Floquet

representation or dressed-molecule picture [1,2]. An important feature of this picture is, in one dimension, an array of curve-crossings occurring among photon-dressed potential energy curves describing multiphoton absorptions. After diagonalization of the radiative interaction, these become avoided crossings. In the vicinity of a given one-photon avoided crossing, the “lower” adiabatic potential presents a barrier to the dissociation. The vibrational dynamics is then governed by shape resonances correlating to field-free vibrational states. The higher the field intensity is, the lower the barrier height will be, and the shorter-lived those shape resonances will become. This is the BS mechanism. VT appears in the same photon-dressed scheme as a complementary mechanism, associated with dynamics that occur through Feshbach resonances supported by the “upper” adiabatic potential which is embedded in, and interacts with, the dissociation continuum of the “lower” one through nonadiabatic couplings.

In a low-frequency, infrared (IR) field, a quasistatic adiabatic picture is appropriate. The vibrational motion develops on the same time scale as the optical cycle. In other words, the important instantaneous distortions of the potential curves follow the field’s oscillations and give rise to a maximally suppressed potential barrier or a maximally closed potential gap as the field periodically reaches its extremum values. The dissociation dynamics depend on the way the motions of the time parametrized adiabatic potential curves distorted by the laser are synchronized with those of the wave packet propagated on them. Efficient DDQ is obtained when this synchronization produces a reflection of the vibrational wave packet on the barrier which is closing as the field is attaining its zero amplitude within the optical cycle [3]. The absolute phase, or carrier-envelope phase [10–12], plays a central role in this synchronization. A DDQ-type effect can also be obtained by synchronizing the vibrational motion, not with the carrier-wave oscillations which may be in the uv-visible range, but with a pulse envelope that is periodically modulated on a time scale corresponding to an IR frequency [4]. The dynamical picture that emerges is an adiabatic photon-dressed potential curve-crossing defined by the uv-visible carrier-wave frequency within the Floquet scheme,

with a “breathing” gap at the avoided crossing, the time dependence of which arising from the slow amplitude modulation of the carrier-wave by the periodic envelope. A DDQ effect of the same type, i.e., one that relies on a synchronization of the dynamics with the pulse envelope, has recently been experimentally observed [8], but it was interpreted in terms of the quasistatic picture as appropriate for an IR carrier wave.

In what follows, we will show how the characteristics of the shape or Feshbach resonances supported by the “lower” and “upper” adiabatic potentials of the uv-visible photon-dressed scheme can dynamically be modified and therefore controlled by the envelope modulations. Our objective is, on one hand, to enhance stabilization by reinforcing VT by DDQ and on the other hand to play against BS with DDQ to overcome dissociation. The opposite objective, i.e., enhancing dissociation by assisting BS with DDQ, could obviously also be considered. The present paper emphasizes the more unexpected enhanced stabilization.

II. MODEL AND METHODOLOGY

We illustrate how the above mechanisms come into play together by considering specifically the stabilization of H_2^+ with respect to its photodissociation in an intense, short laser pulse. This simple model system, a two-state, one-dimensional representation of H_2^+ (supposed to be prepared well-aligned in the linearly polarized laser field), captures the main features of the basic mechanisms. It generically represents a molecule with two electronic states, the ground state $|g\rangle$, ($|g\rangle = |^2\Sigma_g^+\rangle$ for H_2^+), and a dissociative excited state $|u\rangle$, ($|u\rangle = |^2\Sigma_u^+\rangle$ for H_2^+), strongly coupled to each other by the laser electric field $E(t)$ through a transition dipole moment $\mu_{g,u}$. The two electronic states are taken within the Born-Oppenheimer approximation and their energies $\epsilon_{g(u)}$ as well as the transition dipole moment $\mu_{g,u}$ are functions of the nuclear geometry. For the model system considered, these R -dependent quantities are taken from [13]. The complete time-dependent state, including nuclear motions,

$$|\Psi, t\rangle = \chi_g(R, t)|g\rangle + \chi_u(R, t)|u\rangle, \quad (1)$$

is obtained by solving the time-dependent Schrödinger equation,

$$i \frac{\partial}{\partial t} \begin{pmatrix} \chi_g(R, t) \\ \chi_u(R, t) \end{pmatrix} = \left\{ T_N + \begin{pmatrix} \epsilon_g(R) & 0 \\ 0 & \epsilon_u(R) \end{pmatrix} + \mu_{g,u}(R)E(t) \right\} \begin{pmatrix} \chi_g(R, t) \\ \chi_u(R, t) \end{pmatrix}, \quad (2)$$

where T_N is the nuclear kinetic energy operator, using the wave-packet propagation procedure described in previous works [3]. The wave packets $\chi_{g(u)}(R, t)$ are split into two parts: an inner part $\chi_{in,g(u)}(R, t)$, localized in the internal region of the coupled potential energy curves (PECs), where the Hellmann-Feynman forces are important, and an outer part $\chi_{ex,g(u)}(R, t)$, defined in the asymptotic region of the PECs. The inner part is propagated numerically on a grid using the standard third-order split-operator algorithm [14]

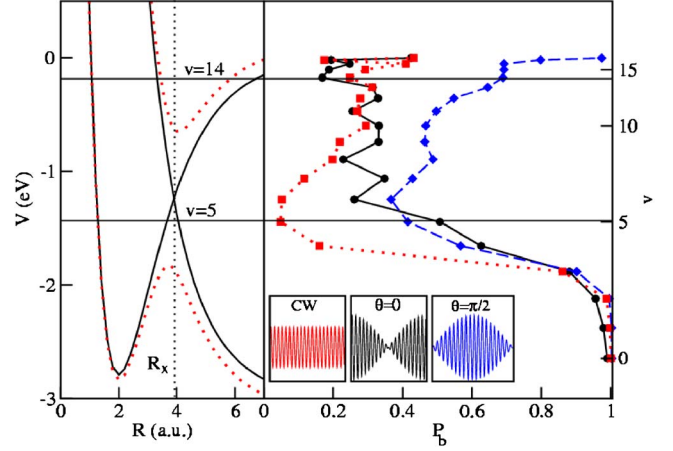


FIG. 1. (Color online) Left panel: Adiabatic (red dotted line) and diabatic (black solid line) dressed potentials of H_2^+ at the one-photon crossing ($\lambda = 2\pi c/\omega = 400$ nm, $I = 2 \times 10^{13}$ W/cm²). Horizontal lines represent the field-free vibrational states $v=5$ and 14 . Right panel: Survival probabilities P_b as a function of the initial state v for the three, equal fluence, pulses [cw (red dotted line), $\theta=0$ (black solid line), $\theta=\pi/2$ (blue dashed line)] represented in the insets.

while the outer part is propagated analytically, by projections onto Volkov-type states [15,16]. Note that by solving Eq. (2) this way, no *ad hoc* hypothesis of an adiabatic time evolution was introduced at the outset, neither in the field-free, nor in the Floquet basis.

We consider a uv-visible carrier field of frequency ω (wavelength λ) modulated by a pulse envelope periodically repeated with a frequency Ω lying in the IR of the form

$$E(t) = E_0 \cos(\Omega t - \theta) \sin(\omega t), \quad (3)$$

θ being the absolute phase of the envelope with respect to which we examine the sensitivity of the dynamics. To better focus on the mechanisms, we further restrict our analysis to a time interval $[0, T_{IR}/2]$, with $T_{IR} = 2\pi/\Omega$ the period of the pulse shape function, i.e., we consider the pulse to be switched off at $t = T_{IR}/2$. Actually, we consider three pulses of equal duration and fluence displayed in the insets (or lower panel) of Fig. 1 and corresponding to a cw field ($\Omega=0$) and to $\theta=0$ and $\pi/2$ pulses. When dressed by the uv-visible photon $\lambda=400$ nm, the potential energy curves for the two electronic states lead to a one-photon avoided curve-crossing in the adiabatic frame which is obtained after diagonalization of the radiative coupling. The left panel of Fig. 1 shows the adiabatic dressed potentials constructed for the cw field of peak intensity of 2×10^{13} W/cm². In order to get a clear signature of the different mechanisms, we consider, for each vibrational level ($|v\rangle, v=0-18$) of the field-free ground electronic state taken as an initial state, the probability for the system to remain bound (right panel of Fig. 1). This is given by

$$P_b(t_f = T_{IR}/2) = \sum_v \frac{|\langle v | \chi_g(t_f) \rangle|^2}{\langle \chi_g(t_f) | \chi_g(t_f) \rangle + \langle \chi_u(t_f) | \chi_u(t_f) \rangle}. \quad (4)$$

The considerations of these vibrational states of the molecular ion as initial states allow one to understand in detail how

the dynamics unfold in terms of individual laser-induced resonances, an understanding which, we hope, will conceptually go beyond the specific system at hand. Moreover, what we show here (for initial v states) should be useful to reconstruct the results expected in the case where a coherent or incoherent sum of v states is required as an initial state, such as in the case one considers the actual preparation of the molecular ion from the parent dihydrogen molecule, by tunnel ionization under the action of the same field, for instance, in which case the initial state, often described as a Franck-Condon wave packet, is a coherent superposition of the molecular ion vibrational states.

Finally, note that insofar as the pulse is switched off at t_f , the quantity P_b defined in Eq. (4) truly represents the total bound state population at any time following the pulse, since in the absence of the field, these bound states cannot decay. No further propagation of the wave packets is needed to calculate P_b , contrary to the computation of an observable that depends on the dissociative state, such as fragment kinetic energy spectra. No such observable is considered in the present paper.

III. RESULTS AND DISCUSSIONS

The results highlight three dissociation regimes: For initial states corresponding to $v \leq 3$, the field-free vibrational states give rise to long-lived shape resonances, well protected against dissociation by the field-induced potential barrier found at the one-photon avoided crossing R_x . The time modulation of the barrier by the pulse envelope function in the case of the two other pulse shapes ($\theta=0$ or $\theta=\pi/2$) does not affect this situation significantly and $P_b(v \leq 3)$ remains larger than 0.85.

Initial states $v=4,5$ correlate with short-lived, light-induced, above-the-barrier shape resonances for which BS is the leading mechanism. This results into very efficient dissociation for the cw field. However, there remains a possibility to quench the dissociation by a proper synchronization between the wave-packet motion and the breathing of the potential gap at the avoided crossing region. This is most clearly seen in the case of the $\theta=0$ pulse with an appropriate modulation frequency Ω . Progressively closing the potential gap by modulating the field amplitude to zero causes a partial inward reflection of the wave packet. This leads to an efficient dissociation quenching, as compared with the situation prevailing in the case of the cw field. Some differences between the cases $\theta=0$ and $\theta=\pi/2$ are also seen although these are not as marked as the ones with the cw field. In the case $\theta=\pi/2$, the gap is opening as the wave packet proceeds towards the dissociation region, and this synchronization normally does not favor a dissociation quenching. However, for such a spatially spread out initial wave packet as the vibrational $v=4,5$ wave functions, the right turning point is so close to R_x that the closed gap seen at the initial time under the $\theta=\pi/2$ pulse already leads to some dissociation quenching, at least more than under the cw field. On this example, we are thus showing that a standard BS mechanism induced by a cw field can be quenched, rather efficiently (by a factor of more than 5, for $v=5$), by referring to a specifi-

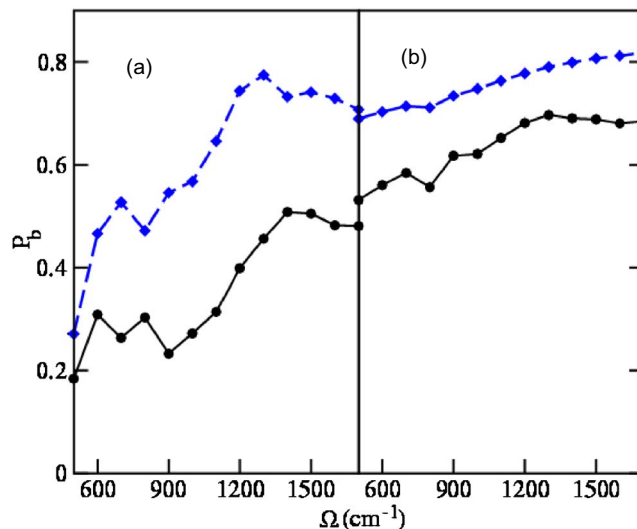


FIG. 2. (Color online) Survival probabilities P_b as a function of the modulation frequency Ω , for two vibrational levels (a) $v=5$ (with $\theta=0$) and (b) $v=14$ (with $\theta=\pi/2$) and peak intensity $5 \times 10^{13} \text{ W/cm}^2$ (black solid line) and 10^{13} W/cm^2 (blue dashed line).

cally shaped pulse. That this is a DDQ-type effect is also evidenced by its sensitivity with respect to the modulation frequency Ω (see also Fig. 2), and to the value of θ .

The states $v \geq 6$ lie above the gap at the one-photon avoided crossing. The vibrational wave packet evolving from them have components both supported by the ground and dissociative potentials. Moreover, the laser field partly transposes the field-free initial vibrational state into dynamical resonance supported by the upper adiabatic potential. This is precisely one of the Feshbach resonances underlying the VT mechanism. As seen in Fig. 1, in this energy range, the survival probability $P_b(v)$ for the cw case tends first (for $v=6-9$) to increase monotonically to finish (for $v > 9$) with an oscillatory behavior about a mean value of 0.2. These oscillations come presumably from further stabilization when an energy coincidence between the initial vibrational level and the resonance position is encountered, a fact well-known in adiabatic/diabatic dynamics [2,17]. It is worthwhile to note that the $\theta=0$ pulse produces similar oscillations about the same mean value of the survival probability, at least for $v \geq 8$. In other words, the $\theta=0$ pulse case does not show any tendency to enhance VT in this energy range. As for the stabilization that this pulse gives for $v=6$ and 7, it arises from the quenching of BS (as for $v=4,5$) rather than from an enhancement of VT. In contrast, at the high energy end, a major quenching effect is observed with the $\theta=\pi/2$ pulse, which dramatically enhances VT with almost full dissociation suppression for the highest vibrational levels.

This DDQ-type reinforcement of VT can be interpreted by a three-step process: (i) Adiabatic transfer. Contrary to the cw and $\theta=0$ pulse cases which start with a sudden switch-on of the field, the $\theta=\pi/2$ pulse, starting from low field amplitudes, permits an efficient adiabatic population transfer between the initial field-free vibrational state and the dynamical Feshbach resonance supported by the upper adiabatic

potential. In such a transfer, only a small wave-packet component is distributed on the lower adiabatic potential and leads to dissociation by BS. The most important component remains the one trapped in the Feshbach resonance. (ii) Feshbach resonance dynamics. Once such resonances are formed, they are exposed, in the case of the $\theta = \pi/2$ pulse, to a field amplitude that is increasing towards a maximum. As this is reached at midpulse, the Feshbach resonance can only decay very slowly into the continuum of the open lower adiabatic channel since the higher the radiative interaction is, the lower the nonadiabatic coupling to this channel will be. A good match between the adiabatic transfer rate and the modulation of the field amplitude of the $\theta = \pi/2$ pulse (as controlled by Ω and E_0), is the key for this DDQ-type enhancement of VT, through a long-lived low-decaying resonance. (iii) Adiabatic transfer. For the $\theta = \pi/2$ pulse under consideration, a second adiabatic transfer occurs at the end of the pulse, between the Feshbach resonance and a field-free vibrational state. The nondissociated population supported by the Feshbach resonance is adiabatically (i.e., with low loss) transferred to a vibrational level of the ground, nondissociative, potential. Here again, only a small wave-packet component is associated with the lower adiabatic channel and can dissociate. This is the first illustration of the possibility of molecular stabilization by combining VT and DDQ mechanisms.

A further support of the interpretations just given can be found in the analysis of the respective roles played by the modulation frequency Ω and the field intensity $I \propto E_0^2$. Figure 2 displays the variations of the survival probability P_b with respect to Ω , for two intensities, namely $I = 10^{13}$ and 5×10^{13} W/cm², and for two initial vibrational states $v = 5$ and 14, representatives of the BS and the VT regimes, respectively. In each case, we focus on the most stabilizing situation, i.e., we consider the $\theta = 0$ pulse for $v = 5$ and the $\theta = \pi/2$ pulse for $v = 14$. The results show that BS is the most sensitive to Ω . This is not surprising as BS relies on the synchronization process controlling the wave-packet transmission versus reflection at the position of the potential gap R_x . VT is less sensitive to the changes in Ω . This is understandable as the dynamics in this case relies on Ω only to ensure a good matching between the rates of the adiabatic transfer of populations from the initial field-free vibrational state and the field amplitude modulation. Otherwise, the stabilization is ensured in this case by the long lifetime of the relevant Feshbach resonance. It is thus the quenching of the still operating BS for the lower adiabatic potential components of the wave packet which imparts the sensitivity of P_b with respect to Ω for $v = 14$, $\theta = \pi/2$ (representative of VT) and a common optimized value leading to the highest bound-state survival probability is found at $\Omega = 1400$ cm⁻¹ in both cases (a) and (b) of Fig. 2. The results for $v = 5$, $\theta = 0$ (representative of BS) are more affected by the varying field intensity than those of $v = 14$, $\theta = \pi/2$. The open potential energy gap necessary for BS to occur is wider for higher field intensity, and increasing the intensity by a factor of 5 leads to a reduction of the survival probability by almost a factor of 2. For $v = 14$, $\theta = \pi/2$, two opposing effects come into competition: On one hand, a stronger field produces a higher rate of dissociation for any component of the wave function

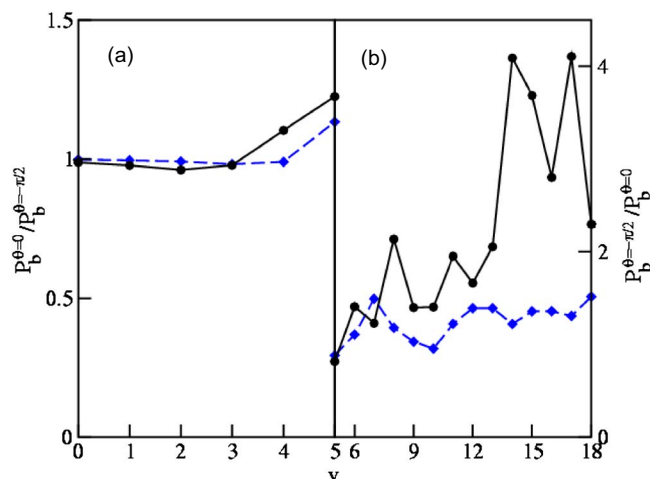


FIG. 3. (Color online) Ratio of survival probabilities (a) $P_b^{\theta=0}/P_b^{\theta=\pi/2}$ or (b) $P_b^{\theta=\pi/2}/P_b^{\theta=0}$ as a function of v , for $\Omega = 1400$ cm⁻¹ and peak intensity 5×10^{13} W/cm² (black solid line) and 10^{13} W/cm² (blue dashed line).

evolving on the lower adiabatic potential, and it may also affect the adiabatic transfers at the beginning and end of the $\theta = \pi/2$ pulse. On the other hand, a stronger field also gives longer-lived Feshbach resonances that are underlying VT. The overall result is slightly dominated by the former effect, a higher intensity still leading to better dissociation. However, for all Ω in the range considered, the ratios of the values of P_b at the higher field intensity with the corresponding ones found at the lower intensity do not exceed 1.25. It is more likely that this behavior arises from the increased dissociation rate of the wave-packet component supported by the lower adiabatic potential rather than from a deterioration of the adiabatic population transfers to/from the stabilized Feshbach resonance. At any intensity, the interplay between the adiabatic transport dynamics (at the beginning and the end of the $\theta = \pi/2$ pulse) and the Feshbach resonance decay is of cooperative nature, resulting into an efficient trapping. Another interesting comparison is obtained by contrasting the results for the two pulsed cases with $\theta = 0$ and $\pi/2$. This is done in Fig. 3, both for the lower v 's of the BS regime, in which case the ratio $P_b^{\theta=0}/P_b^{\theta=\pi/2}$ is plotted as a function of v , and for the higher v 's of the VT regime, for which the inverse ratio $P_b^{\theta=\pi/2}/P_b^{\theta=0}$ is plotted instead, for the two intensities quoted above. While the increasing intensity has a rather modest effect on the distinction between the two pulses in the BS regime (both pulses give rise to significant quenching of BS, as discussed above), it makes the DDQ-enhancement of VT at higher v 's (as obtainable with the $\theta = \pi/2$ pulse) even stronger and $P_b^{\theta=\pi/2}$ is 3 to 4 times larger than $P_b^{\theta=0}$ for $v = 14$ –17 at $I = 5 \times 10^{13}$ W/cm² (this ratio is less than 2 at $I = 10^{13}$ W/cm²), which is a remarkable control result. This observation confirms what was said above, namely, that in the VT regime, a stabilized Feshbach resonance is prepared adiabatically in the case of the $\theta = \pi/2$ pulse and not in the case of the $\theta = 0$ pulse.

IV. CONCLUSIONS

In conclusion, we have shown that efficient control can be exerted on molecular dissociation by shaping a laser pulse in

a relatively simple way. For a field of the form (3), we have illustrated how control can be achieved by adjusting the peak amplitude, the phase θ , and/or the modulation frequency Ω . The underlying understanding is based on the possible enhancement of stabilization, with respect to molecular dissociation, by a dynamical quenching mechanism while inhibiting the BS and reinforcing the VT mechanisms. Since these processes depend on the photon dressed scheme, it is clear that the carrier-wave frequency, [ω in Eq. (3)], will also be an important parameter for the control of the molecular dissociation we are discussing here. Such a control, based on the implementations of well-known basic mechanisms in appropriate combinations, is quite generic and transposable to other systems playing a part in chemical reactivity. Indeed, in a direct extension to a polyatomic molecule or a reactive system, one can imagine how the interplay of these basic mechanisms would operate to control the extent, as well as the outcome, of molecular photodissociation, when a photon-dressed bound potential energy surface (PES) crosses a repulsive PES, both PESs possibly exhibiting multiple dissociative channels. In this respect, we note that the eventual

occurrence of photon-induced conical intersections in polyatomic molecules would present no new conceptual problem, and the generalizations of the above considerations, for example, that of dynamics through time-parametrized, laser-induced Feshbach resonances, remain straightforward. In a less direct generalization, one can apply the same control pattern to nondissociative processes that would merely change the equilibrium geometry of the molecule. An example of this is the interplay of bond-angle hardening (VT-like) and bond-angle softening (BS-like) processes associated with the crossing of the dressed X^2B_1 , \tilde{A}^2A_1 states of the triatomic H_2O^+ molecular ion [18]. These extensions will be explored in future work.

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