PHYSICAL REVIEW A **97**, 033406 (2018)

Control of molecular breakup by an infrared pulse and a femtosecond pulse train

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(Received 9 June 2017; published 16 March 2018)

We investigate the dissociation dynamics of diatomic molecules subjected to both a femtosecond infrared (IR) laser pulse and a femtosecond pulse train (FPT) within the framework of the Morse potential model. When the IR and FPT are phase delayed, we observe well-resolved oscillations in dissociation probability, corresponding to multiple integers of the IR period, exhibiting enhancement and suppression of bond dissociation. These oscillations reveal a rich dynamics as a function of the IR and FPT parameters including chaotic fields. A frequency-resolved profile of dressed molecular states shows that these oscillations are due to interference of many quantum paths analogous to the recently observed control of photoionization of atoms under IR and XUV pulses. By manipulating phases of FPTs we demonstrate an enhancement of molecular dissociation compared to the transform-limited case.

DOI: 10.1103/PhysRevA.97.033406

I. INTRODUCTION

The dissociation of a molecular bond by light is a fundamental phenomenon with important applications in controlling various physical, chemical, and biological processes [1,2]. However, the anharmonicity of the molecular potential energy makes it difficult for a single weak infrared (IR) laser pulse to efficiently dissociate a molecule. For strong pulses, molecular tunnel ionization often dominates over dissociation [3–5]. Many different approaches based on coherent and/or incoherent excitation have been proposed to enhance the photodissociation. For example, the use of linearly (or circularly) polarized chirped laser pulses facilitates the ladder climbing of a chosen molecule and eventually its dissociation [6], which has been also confirmed experimentally with vibrational excitation observed in diatomic (HCl, NO, etc.) and polyatomic molecules ($Cr(CO)_6$) [7–11]. The optimal pulse shaping of a femtosecond pulse, along with closed-loop control, has been used to control the branching ratio between two possible reaction channels [12].

Photodissociation of small molecules was studied using a variety of methods, such as the combination of an intense IR laser with a weak UV laser for dissociating HOD [13,14], or optimally shaped pulses with five lasers [15] and using a single intense XUV pulse [16]. A single-cycle terahertz pulse has been shown to achieve ladder climbing on anharmonic amino acid crystals up to the 20th vibrational state [17]. Recently, attosecond control of molecular dissociation has been demonstrated by means of attosecond recollision [18], attosecond molecular photoionization [19], electron localization [20], and subfemtosecond steering of protons through superposition of vibrational modes [21].

A completely different approach has been proposed to incoherently control the molecular photodissociation by subjecting it to incoherent random kicks [22–26]. The molecular dissociation processes due to the simultaneous application of a femtosecond laser pulse and a noise source has been shown to enhance the dissociation analogous to the quantum stochastic resonance phenomena [27–29]. While most of these approaches enhance molecular dissociation, only few approaches offer good control, i.e., both enhancement and suppression of dissociation by varying a control parameter.

Here, we generalize the idea of attosecond control of atomic photoionization by XUV and IR pulses [30] to the dissociation dynamics of small molecules. This leads to an alternative route to control molecular dissociation by irradiating a diatomic molecule with a mid-IR femtosecond pulse along with a femtosecond pulse train (FPT), which is synthesized by combining high-order harmonics of the mid-IR pulse [31–34]. We show results on the dissociation dynamics of the molecule subjected to different FPTs with phase-locked or phase-randomized harmonics by numerically solving the time-dependent Schrödinger equation. Our approach is applicable to both polar and nonpolar molecules and is suitable for experiments.

The paper is organized as follows. In Sec. II, we briefly describe our model, which is a Morse potential driven by a coherent mid-IR laser pulse and a FPT. Section III shows our results for the control of molecular dissociation using a phase-delayed action of FPT and IR pulse. We demonstrate a rich dynamics of coherent oscillations in the dissociation probability by varying model parameters. In particular, an enhancement of the yield by randomizing only phases of the harmonics is demonstrated. Finally, Sec. IV concludes the paper with some perspectives.

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2469-9926/2018/97(3)/033406(6)

II. MODEL

A. Driven Morse potential

We choose a generic Morse potential model of molecular vibrations which is subjected to a femtosecond mid-IR laser pulse and a FPT. The Hamiltonian for the system is given as (atomic units are used unless stated otherwise)

$$H(t) = H_0 - xF(t) - xF_{pt}(t), \tag{1}$$

where the molecular dipole gradient [22] has been absorbed into the external perturbations $F_{pt}(t)$ and $F(t)$ to the molecule. The Hamiltonian $H_0 = \frac{p^2}{2m} + V(x)$ describes vibrational motion of the molecule of the reduced mass m in the Morse potential [35],

$$V(x) = -D_e + D_e[1 - \exp(-\beta x)]^2, \tag{2}$$

with well depth D_e and length scale β. The eigenenergies E_n of the Morse oscillator H_0 are given by

$$E_n = \hbar\omega_e(n + 1/2)[1 - B(n + 1/2)/2], \quad 0 \leqslant n \leqslant [j], \tag{3}$$

where ω_e is the harmonic frequency and $n_b = [j] + 1$ is the number of bound states with

$$j = 1/B - 1/2, \quad B = \hbar\beta(2mD_e)^{-1/2}, \quad \hbar\omega_e = 2BD_e. \tag{4}$$

The Morse potential as stated above can be applied to both polar and nonpolar diatomic molecules as well as to a particular bond in some complex polyatomic molecules [7,8,11].

In the following, we choose the specific example of a HF molecule ($D_e = 6.125$ eV, $\beta = 1.1741$, and $m = 1790.35$), which has 24 bound states. The vibrational ground-state energy is -0.21 a.u. and the ground to first vibrational excitation corresponds to 0.01807 a.u. [22,24]. The mid-IR driving frequency is chosen much smaller and is nonresonant to lower vibrational states. The molecule is considered oriented along the laser polarization axis and for this mid-IR driving the ionization in the molecule is considered negligible [22,36].

The laser field is a nonresonant mid-IR femtosecond pulse with time-dependent amplitude,

$$F(t) = f(t)F_0 \sin(\omega t + \phi). \tag{5}$$

Here F_0 denotes the peak amplitude of the pulse, ω denotes the angular frequency, and ϕ is the carrier envelope (CE) phase. We choose a smooth pulse envelope $f(t)$ of the form

$$f(t) = \sin^2(\pi t/T_p), \tag{6}$$

with $f(t) = 0$ for $t < 0, t > T_p$. Here T_p is the pulse duration of typically ten optical cycles [see Figs. 1(a1) and 1(a2), in green].

The femtosecond pulse train is defined as

$$F_{pt}(t) = f'(t)\frac{F_{0pt}}{N}\sum_{n=n_i}^{n=N_f} \sin(\omega_n t + \phi_n), \tag{7}$$

where F_{0pt} is the peak amplitude of the FPT pulse, and ω_n and ϕ_n are the frequencies and phases of the nth harmonic, respectively. The pulse envelope $f'(t)$ is chosen identical to that of the IR pulse in Eq. (6). We synthesize two types of FPT from odd-order harmonics and all (even plus odd) harmonics of the driving field. Notice that the pulse emerging from the FPTs can be either coherent or incoherent (chaotic), depending on the choice of the phases ϕ_n. In a special case, when all the phases are locked to, say, zero (i.e., $\phi_n = 0$), the FPT is Fourier transform limited (FTL) with the shortest possible femtosecond bursts [see, e.g., Figs. 1(a1) and 1(a2), in blue]. However, when theses phases are randomly assigned, the pulse may instead look chaotic, due to random superposition of the harmonics [24].

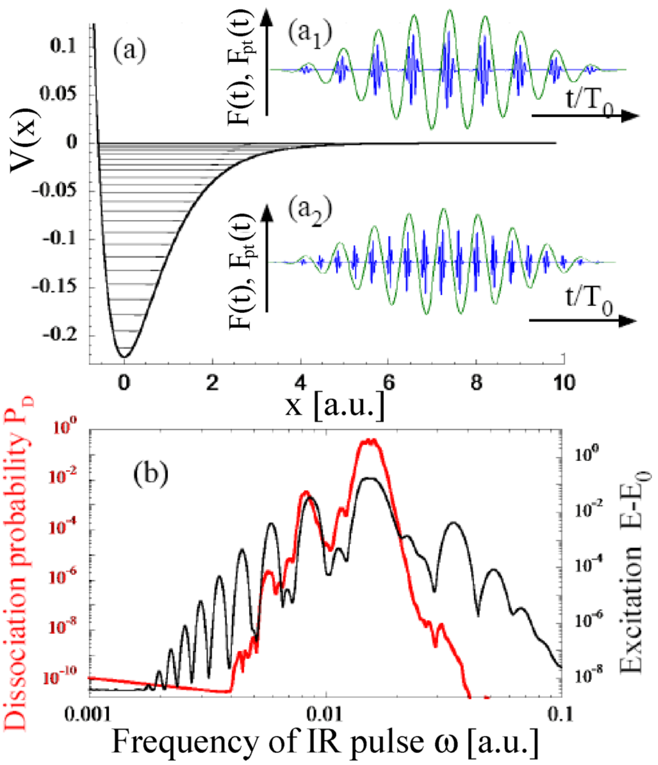

FIG. 1. The picture of Morse potential $V(x)$ for a diatomic molecule HF. Here $x = R - R_0$ is relative displacement from the equilibrium. Mid-IR pulse (green) and two cases of FPT (blue) with (a1) one pulse or (a2) two pulses per cycle of mid-IR pulse. The simulations correspond to total $N = 9$ harmonics (a1) from $N_i = 5$ to $N_f = 13$ and (a2) from $N_i = 5$ to $N_f = 21$. The mid-IR pulse is ten cycles long with an angular frequency $\omega = 0.007$ and CE phase $\phi = 0.2$. (b) Dissociation probability P_D (red) and excitation from the ground state (black) as a function of ω when the molecule is driven by an IR pulse only. The multiple peaks for ω below 0.01 show multiphoton excitation of the molecule from its initial ground state.

B. Dynamics and observables

The dynamics of our system as described above is in general governed by the time-dependent Schrödinger equation (TDSE) of the form

$$i\hbar\frac{\partial|\psi(t)\rangle}{\partial t} = H(t)|\psi(t)\rangle\,, \tag{8}$$

for which a solution can be efficiently obtained using the fast Fourier transform split-operator method with absorbing boundary conditions [23,37]. The nature of this dynamics is dictated by the type of FPT. If the pulse originating from the FPT is coherent, the dynamics is deterministic and relies upon the solution of the standard TDSE. However, if this pulse shape is chaotic, each random sequence of fixed phases constitutes one realization of the noise in the FPT and its solution may require averaging over many realizations of the noise.

The diatomic molecule is considered to be initially in the ground vibrational state $|\psi_0\rangle \equiv |0\rangle$ with energy E_0 [Eq. (3)]. The ground state is analytically known in this case and can be written as [35]

$$|\psi(0)\rangle = N_0 \exp(-f(x)/2) f(x)^j, \qquad (9)$$

where $f(x) = k \exp(-\beta x)$ and N_0 is the normalization constant.

In order to characterize the nonlinear interplay between the laser pulse and the FPT, we mainly compute the dissociation probability P_D, which quantifies the amount of population in the continuum states. It is easier to calculate the complement, i.e., the population of the bound state ψ_ν, with

$$P_\nu(t) = |\langle\psi_\nu|\psi(t)\rangle|^2. \qquad (10)$$

The dissociation probability for a realization is then given by

$$P_D(t) = 1 - \sum_{\nu=0}^{N_b-1} P_\nu(t). \qquad (11)$$

Typically, for a given realization we calculate the dissociation probability $P_D(t)$ over a long time $t_\infty \gg T_p$. We compute $P_D(t_\infty)$ for various parameters of the model including the CE phase ϕ and two types of FPT, synthesized from odd-order or all harmonics, with their phases ϕ_n locked either to zero or to random values.

III. RESULTS AND DISCUSSIONS

A. Oscillations in the photodissociation probability

We first compute the dissociation probability P_D when the molecule is driven by intense IR pulse only as a function of the pulse frequency ω [Fig. 1(b)]. A similar response in the dissociation of H_2^+, in particular, near unity dissociation for pulse frequencies around 0.02 has been previously reported [36], which corroborates the validity of our model. However, to demonstrate control of molecular dissociation we choose a weak mid-IR pulse, at central frequency $\omega = 0.007$ and peak amplitude $F_0 = 0.03$, that produces a small dissociation probability ($P_D < 10^{-7}$). We overlap such a weak mid-IR pulse with a FPT composed of either (i) odd harmonics only or (ii) both even and odd harmonics. Both kinds of FPT have been experimentally synthesized using one- or two-color driving fields [30,38]. Since we focus on the sub-IR-cycle oscillation regime, we overlap the IR with FPT pulses and vary the CE phase ϕ of the IR pulse. In our case, varying ϕ, for our ten-cycle-long pulse, is equivalent to changing the time delay in one optical cycle between IR and FPT.

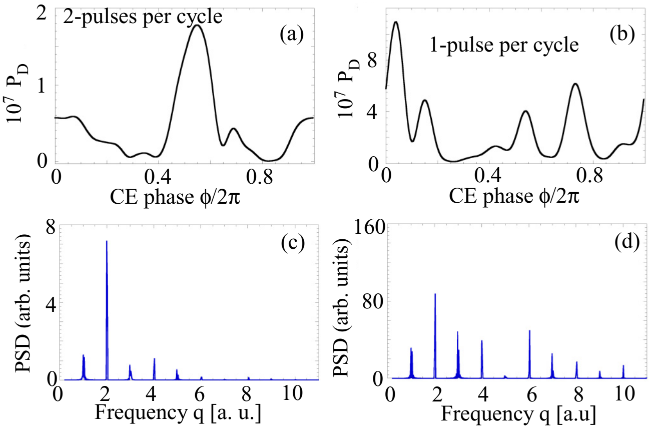

FIG. 2. The dissociation probability P_D for two different cases of FPT containing (a) two pulses per cycle (odd harmonics) and (b) one pulse per cycle (all harmonics). (c, d) Corresponding power spectral density computed from Fourier transform of P_D over a quasiperiodic function over many 2π CE phases. The frequency q is a dimensionless frequency defined as $2\pi/\phi$ and it is not related to the mid-IR frequency. Here, $F_0 = 0.04$, $F_{0pt} = 0.1$, and the HH spectra consist of $N = 9$ harmonics.

Figures 2(a) and 2(b) show the dissociation probability P_D as a function of ϕ for both cases of odd-only and even-plus-odd harmonics, respectively. P_D exhibits a structure that varies by more than two orders of magnitude within an optical cycle as the CE phase is varied from zero to 2π. In order to reveal the spectral contents in the oscillations of $P_D(\phi)$, we perform its Fourier transform $P_f(\Omega)$ [see Figs. 2(c) and 2(d)]. To calculate the fast Fourier transform (FFT), we replicate this signal $P_D(\phi)$ over several tens of cycles and then take the Fourier transform, $P_f(q) = \int P_D(\phi) \exp(-iq\phi) d\phi$. The FFT frequency q is a dimensionless variable which has a representation of $2\pi/\phi$ and is not related to the IR frequency. Alternatively, one can also computes P_D versus the time delay (for fixed CE phase) as was performed in atomic cases in Refs. [30,39], where one can consider $\phi(t) = \omega t$ where t is some arbitrary parameter having the same dimension as time. In the cases of odd-only and even-plus-odd harmonics, the power spectral density (PSD) of oscillations show well-resolved components $q = 1,2,3,\ldots$. In fact, in all our simulations for various values of F_0, fundamental frequency, and FPTs, P_D contains only integer values of q.

We further isolated different specific frequencies present in the observed photodissociation P_D by "filtering" the harmonics spectra and computing the dissociation response to different pairs of harmonics. In Fig. 3(a) we show the dissociation probability for four possible harmonics pairs separated by twice the fundamental frequency, i.e., when Eq. (5) contains H7+H9, H9+H11, H11+H13, and H13+H15. In these cases, we observe a dominant periodicity of twice the fundamental frequency. These equal-frequency oscillations arise due to interference between two-photon absorption and emission involving one high-order harmonic and mid-IR photon, as depicted in Fig. 3(d). Similarly, when we choose the harmonic pairs separated by four or six times the mid-IR frequency, we observe oscillations in the photodissociation probability at four and six times the fundamental frequency, respectively [Figs. 3(b) and 3(c)]. They are due to interference between quantum paths corresponding to absorption and emission of two and three IR photons following absorption of high-order-harmonic photons as depicted in Fig. 3(d). When all the harmonics are present simultaneously, these multiphoton paths are simultaneously excited and superposition of all interferences gives rise to structure in the dissociation probability as a function of the CE phase.

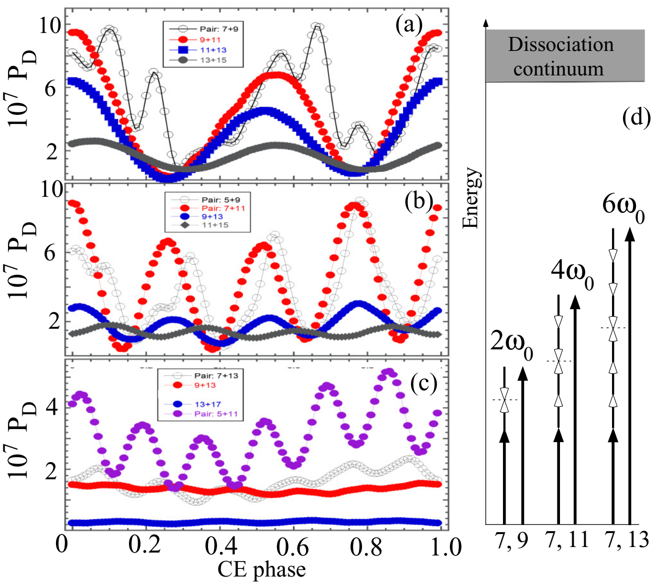

FIG. 3. Photodissociation probability versus CE phase when the harmonics are chosen in pairs, separated by (a) twice the fundamental frequency, (b) four times the fundamental frequency, and (c) six times the fundamental frequency. (d) Schematic representation of the corresponding quantum path interferences exciting the molecule from the initial state to final states by absorbing or emitting multiple photons.

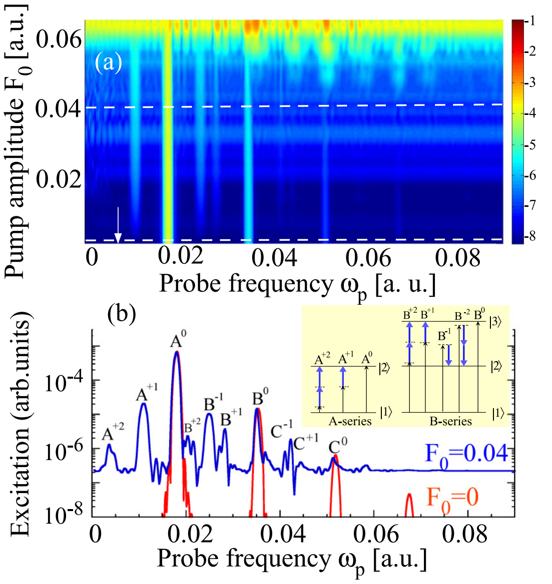

FIG. 4. (a) The frequency-resolved absorption profile of a HF molecule dressed with a mid-IR pulse of fixed frequency $\omega_0 = 0.007$ and variable amplitude. The excitation of the molecule is measured with a second pulse variable frequency ω_p and weak amplitude, $F_0/F_{0pt} = 20$. (b) Cuts taken along the dashed line at $F_0 = 0.04$ and comparison with the unperturbed spectrum at $F_0 = 0$. Without the mid-IR pulse, the peaks (in red) correspond to one-photon vibrational transitions. With the mid-IR pulse, multiple peaks (in blue) correspond to excitation or deexcitation of molecules by simultaneous absorption and emission of multiple mid-IR photons. The inset shows the corresponding multiphoton paths for the first two excited states.

To understand the mechanism behind the oscillations in P_D, we measured a frequency-resolved absorption profile of the dressed molecule. The dressing field was the mid-IR pulse at fixed frequency $\omega = 0.007$ but of different amplitudes F_0. To probe the dressed states we computed absorption of another weak laser pulse of variable frequency ω_p. The tunable pulse has the same pulse duration and shape. The amplitude of this pulse is much weaker ($F_0/F_{0pt} = 20$), such that it produces mostly single-photon transitions and its frequency is varied from a very small value of 0.001 to the maximum frequency that corresponds to a direct one-photon transition to the vibrational continuum. The absorption profile of the dressed molecule, initially in its ground state, was obtained by measuring the net energy absorbed ($E - E_0$) as a function of ω_p and amplitude F_0 as shown in Fig. 4. The absorption profile of the dressed molecule exhibits several distinctly resolved peaks. It turns out that for our FPT consisting of odd harmonics, from $n = 5$ to $n = 21$, the lower-energy harmonic components play a dominant role in the enhancement process.

The coherent oscillations in the dissociation probability results from an interference among multiphoton paths due to simultaneous absorption of mid-IR and high-order harmonics (HH) photons. The various possible multiphoton paths vibrationally excite the molecule which has been quantified by computing the expectation value of the change in total energy of the molecule, $E - E_n$, from the nth initial state. In particular, for a molecule in the ground vibrational state, $E - E_0$ is always positive [Fig. 1(b)], i.e., the molecule is only entitled to absorb the energy. However, this quantity can be either positive or negative for an initially excited molecule which can climb up or down the ladder, respectively. Figure 5 shows the change in energy of the molecule from $n = 2$ and $n = 6$ states as a function of the frequency. This shows that the vibrational excitation as well as deexcitation takes place through distinct paths corresponding to one-, two-, three- and more-photon processes simultaneously. The dressed states depend on the amplitude and frequency of the IR pulse, and for large enough

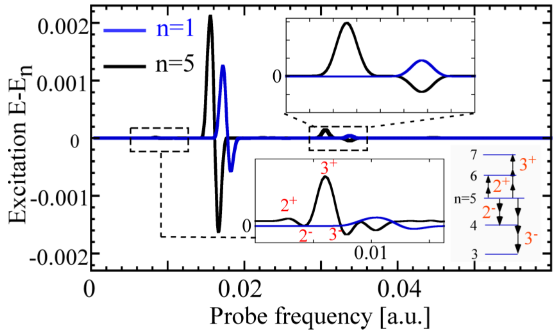

FIG. 5. The excitation of the molecule versus probe frequency ω_p when it was initially in the $n = 1$ (first excited state) and $n = 5$ (fifth excited state) vibrational state. The peaks and dips (i.e., excitation and deexcitation) correspond to the transition between vibrational states of energies $E_{n+1} - E_n$ and $E_n - E_{n-1}$. The dip in the blue curve in the upper inset is missing because the deexcitation below the ground state is not possible. Lower inset: excitation and deexcitation by absorption and emission of multiple photons with corresponding paths labeled.

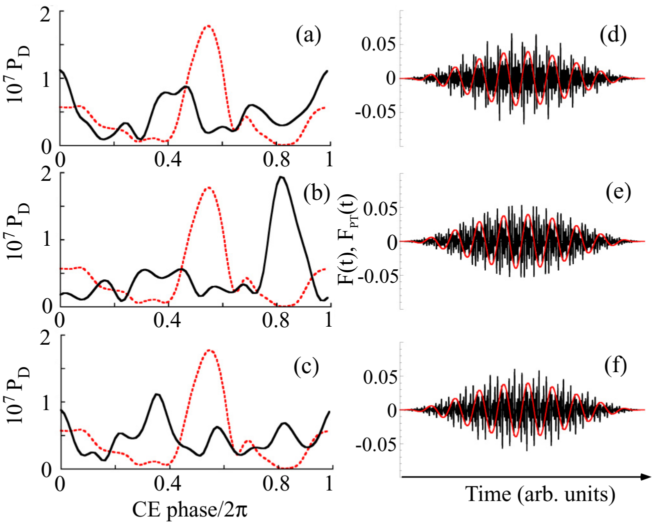

FIG. 6. (a)–(c) Typical dissociation probability versus CE phase for three different realizations of the chaotic FPT. (d)–(f) The chaotic waveforms are generated randomly, assigning the phases ϕ_n from zero to 2π, respectively. The dotted line shows the dissociation probability P_D versus CE phase corresponding to the transform-limited pulses corresponding to Fig. 2(a).

amplitude ($F_0 > 0.05$) neighboring states tend to overlap each other and become difficult to interpret.

B. Photodissociation enhancement for phase-randomized FPT

We have seen in Figs. 4 and 5 that the vibrational (de)excitation sensitively depends on the parameters of the mid-IR pulse as well as on the HH frequency components of the FPT. In addition, we demonstrate in Fig. 6 that, for a given spectral content of the FPT, the oscillation in P_D can also be controlled by phases ϕ_n of the harmonics. We show different realizations of phase-randomized FPT along with the superimposed IR pulse (in red). The corresponding dissociation yield versus CE phase ϕ is also shown. When compared with the transform-limited FPT, the chaotic-like FPT can enhance the contrast of the oscillations. This demonstrates that the sensitivity of the interference of multiphoton paths to the phases ϕ_n of the HH can be exploited to improve the degree of control, suggesting a possible mode-locking-type synchronization of frequency components in P_D. Figure 6 shows that, in the case of a FTL pulse train, producing the shortest pulse may not be ideal to achieve maximal control. However, if we choose a pulse that looks chaotic, it may produce an even more enhanced contrast of the oscillations.

IV. SUMMARY AND CONCLUSION

In summary, we have demonstrated significant control of bond breaking using a mid-IR pulse and a femtosecond pulse train composed of its high-order harmonics. The Morse potential model is numerically solved within the framework of the time-dependent Schrödinger equation. For overlapping IR with FPT, we observe well-resolved oscillations in the dissociation probability as a function of the CE phase of the IR. The oscillations in dissociation probability are generic features of both types of FPT composed of either odd-order or all-order harmonics. The rich dynamics in the strong-field oscillations is a result of quantum interference among various multiphoton paths to molecular excitation.

This approach is analogous to coherent control of photoionization of atoms observed with an IR pulse and an attosecond pulse train. One may potentially synchronize various spectral components of dissociation channels to further improve the control of the dissociation probability. In this direction, it could be worthwhile to implement a genetic-algorithm-based optimization approach to further obtain the maximum possible control. While the proposed approach can yield significant control for small diatomic molecules (H_2, HCl, etc.), it would be of interest to study the interplay between an IR pulse and FPT on selective dissociation and ionization in polyatomic molecules. Our results might be relevant to situations where more complex molecules, quantum dots, or nanoparticles are exposed to a mid-IR pulse (acting as a photonic catalyst) to control excitations in other quantum systems such as in quantum dots [40], or in vibrationally promoted dissociation of water [41,42].

ACKNOWLEDGMENTS

K.P.S. acknowledges financial support from the Max Planck Society Germany and DST New Delhi.

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