Switched Wave Packets: A Route to Nonperturbative Quantum Control

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The dynamic Stark effect due to a strong nonresonant but nonionizing laser field provides a route to quantum control via the creation of novel superposition states. We consider the creation of a field-free "switched" wave packet through adiabatic turn-on and sudden turn-off of a strong dynamic Stark interaction. There are two limiting cases for such wave packets. The first is a Raman-type coupling, illustrated by the creation of field-free molecular axis alignment. An experimental demonstration is given. The second case is that of dipole-type coupling, illustrated by the creation of charge localization in an array of quantum wells.

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The development of coherent laser sources and the control over optical phase implied has emerged as an important tool for the control of quantum processes. Brumer and Shapiro as well as Tannor and Rice developed coherent optical phase control schemes based on the quantum interference between two or more pathways to a degenerate target state [1]. Although these coherent control schemes are often based on a perturbative description in that the eigenstates of relevance are those of the field-free Hamiltonian, schemes using strong resonant or near-resonant laser fields have also been developed [2]. An alternative approach based on the nonlinear interaction of an adaptively shaped laser pulse using feedback-learning algorithms to optimize a chosen product was proposed by Rabitz [3]. This was most successfully demonstrated in the strong field fragmentation ionization of polyatomic molecules [4] and atomic ionization [5]. These schemes are nonperturbative in that the field-free Hamiltonian may not be pertinent, and they may take advantage of many processes including multiphoton resonances, dynamic Stark effects, propagation on multiple potential energy surfaces, and strong field ionization-fragmentation processes [6]. In nonionizing laser fields, strong shaped pulses have been used to control processes in condensed phases [7] and may take advantage of multiphoton resonances and intrapulse interferences [8] and dynamic Stark shifts. Although the underlying mechanisms are complex and difficult to elucidate, strong field shaped pulse feedback control has been as successful as coherent quantum interference control. Here we consider nonperturbative control schemes which are restricted *only* to the dynamic Stark effect induced by nonresonant, nonionizing laser fields. By choice of laser frequency and intensity, complex competing processes might be eliminated.

As a first step to this nonperturbative control, we study experimentally and theoretically the creation of field-free ground state superpositions via the adiabatic application of a strong nonresonant dynamic Stark coupling which is suddenly truncated, thus forming a wave packet. This "switched wave packet" will be very different in nature and composition than that created via resonant excitation or strong field ionization. We consider the interaction of a strong nonresonant laser field of the form $\mathbf{E}(t) = \hat{\mathbf{e}}\mathcal{E}(t)\cos(\omega t)$ with a quantum system, where $\hat{\mathbf{e}}$ is the direction of polarization of the electric field of frequency ω with a slowly varying envelope $\mathcal{E}(t)$. The Hamiltonian of the system in the presence of the laser field is then $H(t) = H_0 - \mathbf{d}\cdot\mathbf{E}(t)$, where H_0 is the field-free Hamiltonian and $\mathbf{d}$ is the dipole moment of the system. The field-free eigenstates can often be separated into bands, as depicted in Fig. 1, representing the energy bands of a molecule, a set of quantum wells, a Stark or Zeeman manifold, or some other structure. If the optical frequency is greater than δ and smaller than Δ, the dynamics may be represented by an effective Hamiltonian where all other bands have been adiabatically eliminated. The equations of motion for the wave function $\Psi(t) = \sum_n a_n(t)e^{-iE_n t}|n\rangle$ expressed as a superposition of the initially populated band ($N = 0$) eigenstates are [9]

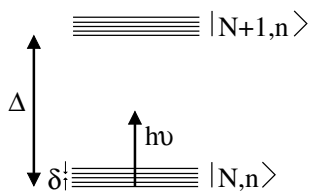

FIG. 1. A quantum system described by quantum numbers N and n. The laser frequency is chosen to be much larger than the intraband spacing δ (described by n) and much smaller than the interband spacing Δ (described by the N).

$$i\dot{a}_n(t) = \sum_m a_m(t) e^{i\omega_{nm}t} H^{\text{eff}}_{nm}(t), \quad (1)$$

where the leading terms of the effective Hamiltonian are

$$H^{\text{eff}}(t) = V^{\text{dipole}}(t) + V^{\text{Raman}}(t), \quad (2)$$

$$V^{\text{dipole}}_{nm}(t) = -\mathcal{E}(t)\cos(\omega t)\hat{\mathbf{e}} \cdot \mathbf{d}_{nm}, \quad (3)$$

$$V^{\text{Raman}}_{nm}(t) = -\tfrac{1}{4}|\mathcal{E}(t)|^2 \hat{\mathbf{e}}^* \cdot \boldsymbol{\alpha}_{nm}(\omega) \cdot \hat{\mathbf{e}}, \quad (4)$$

where $\mathbf{d}_{nm} = \mathbf{d}_{0n0m}$ is the transition dipole moment and

$$\boldsymbol{\alpha}_{nm}(\omega) = \sum_{N\neq 0}\sum_i \mathbf{d}_{0n\text{Ni}}\mathbf{d}_{\text{Ni}0m}\left[\frac{1}{\omega_{\text{Ni}} + \omega} + \frac{1}{\omega_{\text{Ni}} - \omega}\right]. \quad (5)$$

In the situation where the intraband dipole moments are large, $V^{\text{dipole}}_{nm}(t)$ will dominate and the response follows the instantaneous electric field. Conversely, when the interband dipole moments are large, $V^{\text{Raman}}_{nm}(t)$ dominates and the response follows the electric field envelope $|\mathcal{E}(t)|^2$. These situations represent the two limiting cases for switched wave packets, and an example of each will be presented below.

When the interaction is turned on adiabatically [10], each eigenstate of the field-free Hamiltonian evolves into a single instantaneous eigenstate of the laser dressed system. The accumulated phase is given by the time integral of the instantaneous eigenenergy. Adiabatic behavior is achieved through sufficiently slow turn-on of the laser field and limiting the intensity to avoid crossing of the dressed states [11]. For the limiting case of a Raman dominated interaction, the application of the adiabatic approximation is clear and the relevant states and quasienergies are easily found from Eq. (4) [11]. For the case of a dipole dominated interaction, the Floquet states which explicitly include the fast electric field oscillations provide a useful description [12]. The adiabatic condition here refers to varying the envelope and carrier frequency slowly enough to avoid transferring population between the Floquet states. In either limiting case, a nonadiabatic change in the interaction leads to a superposition of dressed states and beats at frequencies corresponding to the differences between quasienergies of the dressed states [10]. This results in Rabi oscillations in the field-free state populations. A sudden truncation of the field will project the dressed state onto the field-free states forming a wave packet which subsequently evolves under the field-free Hamiltonian. The content of this wave packet will depend upon the nature of the strong interaction, the turn-on time τ_{on}, and the switching time τ_{sw}.

We first consider the limiting case of a switched wave packet in which the interaction is dominated by the Raman term, Eq. (4), an example being molecular rotors. An anisotropic linear molecule placed in a linearly polarized laser field experiences a potential of the form [13]

$$V(\theta, t) = -[\tfrac{1}{4}(\alpha_\parallel - \alpha_\perp)|\mathcal{E}(t)|^2]\cos^2(\theta), \quad (6)$$

where θ is the angle between the laser polarization and the molecular axis, $\alpha_\parallel$ and $\alpha_\perp$ are the polarizability components parallel and perpendicular to the molecular axis, and the term in square brackets corresponds to the well depth $U(t)$. The Hamiltonian is then $H = B\mathbf{J}^2 + V(\theta, t)$, where $\mathbf{J}$ is the angular momentum operator and B is the molecular rotational constant. The aligned (pendular) molecule is described by a superposition of field-free rotational eigenstates $\psi(t) = \sum a_{JM}|JM\rangle$. Since $V(\theta, t)$ is independent of ϕ, the Hamiltonian is separable in θ and ϕ, implying conservation of M. We consider here a pulse shape of the form

$$|\mathcal{E}(t)|^2 = \begin{cases} \mathcal{E}_0^2 \sin^2\left(\frac{\pi t}{2\tau_{\text{on}}}\right), & t \le \tau_{\text{on}}, \\ \mathcal{E}_0^2 \cos^2\left(\frac{\pi(t-\tau_{\text{on}})}{2\tau_{sw}}\right), & \tau_{\text{on}} < t \le \tau_{\text{on}} + \tau_{sw}. \end{cases} \quad (7)$$

For each initial J, a single pendular state will be created if τ_{on} is long enough for adiabatic behavior. If τ_{on} is too short, the state created will be a superposition of pendular eigenstates and the wave packet will have additional time dependence to its alignment [13]. A switched rotational wave packet calculation is depicted in Fig. 2. A strong field pulse with $\tau_{\text{on}} = 15$ ps was used to create a switched wave packet in N_2 at 50 K. Alignment is measured by the expectation value of $\cos^2(\theta)$. At time t_{sw}, the pendular state is suddenly projected with $\tau_{sw} = 0.1$ ps onto the free rotor states, forming a field-free rotational wave packet. We present an experimental demonstration of a switched rotational wave packet. Briefly, a 1.064 μm laser pulse with τ_{on} of 125 ps and τ_{sw} of $\sim$100 fs was generated using a plasma shutter method. A femtosecond Ti:Sa oscillator (80 fs, 800 nm) and a picosecond Nd:YAG oscillator (125 ps, 1.064 μm) were electronically synchronized with a timing jitter of $\sim$1 ps. Both oscillator pulses were amplified at a repetition rate of 1 kHz. A two stage Ti:Sa amplifier system produced 2.5 mJ, 80 fs pulses. The

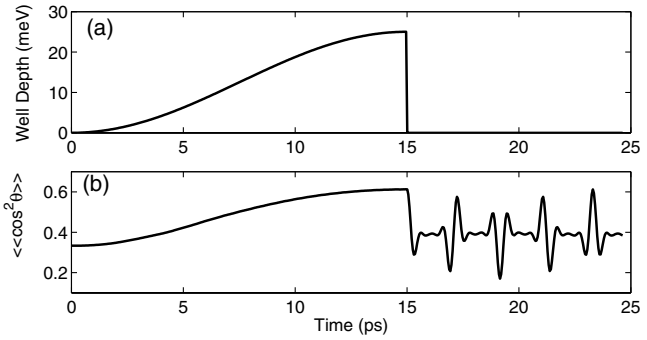

FIG. 2. Switched rotational wave packet in N_2 at 50 K. (a) The well depth, $U(t)$, created by a laser field with $\tau_{\text{on}} = 15$ ps and $\tau_{sw} = 0.1$ ps. (b) The time-dependent alignment of the ensemble showing wave packet revivals and field-free alignment.

picosecond amplifier was a diode pumped Nd:YVO_4 laser [14] producing 1 mJ, 125 ps pulses. The 1.064 μm beam was focused into an ethylene glycol jet with an intensity of $\sim 5 \times 10^{11}$ W cm^{-2}, below the threshold for breakdown. A fraction (600 μJ) of the 800 nm beam was focused to the same spot position and size in the jet with a peak intensity of $\sim 5 \times 10^{14}$ W cm^{-2}, well above the threshold for breakdown. The 800 nm triggered plasma acted as a shutter for the 1.064 μm pulse, truncating its transmission. In our case, the plasma was sustained by the intense 1.064 μm pulse. The transmitted 1.064 μm light had $\tau_{\text{on}} = 125$ ps and $\tau_{sw} \sim 100$ fs, as seen in Fig. 3(a). Residual scattered 800 nm light copropagating with the 1.064 μm pulse was removed by dichroic beam splitters.

The 1.064 μm switching pulse was focused into a gas cell containing CO_2 (300 torr) with peak intensity of $\sim 10^{11}$ W cm^{-2} corresponding to a well depth of ~ 0.7 cm^{-1}. The alignment induced by the switching pulse was probed via optical Kerr effect (OKE) rotation of a copropagating weak 80 fs probe pulse, derived from the remainder of the 800 nm 80 fs pulse. Prior to the gas cell, this probe passed through a Glan-Taylor polarizer oriented at +45° clockwise with respect to the switching pulse. The 800 nm probe energy transmitted through a second crossed polarizer ($-45°$) was recorded as a function of time delay between the switching and probe pulses ($\Delta t = t_{\text{probe}} - \tau_{sw}$). The extinction ratio of the crossed polarizer pair was $<10^{-6}$. The transmitted probe pulse was dispersed through a 0.4 m monochromator and detected with a photomultiplier tube.

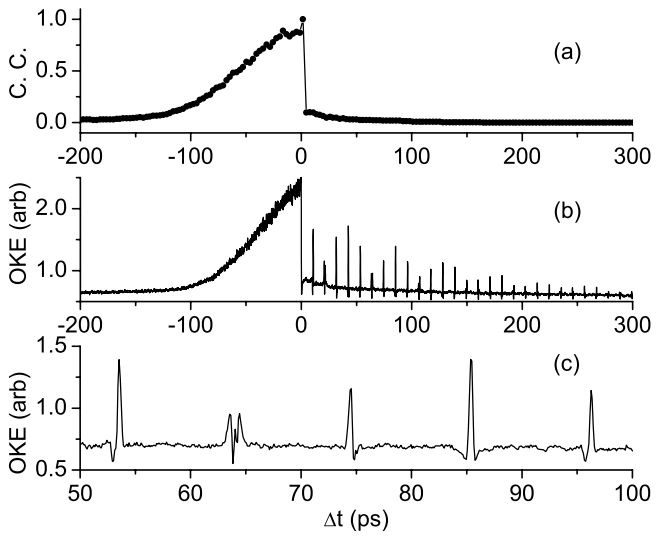

FIG. 3. Experimental demonstration of switched wave packets. (a) Cross correlation (C.C.) of the switching 1.064 μm laser pulse with an 80 fs, 800 nm pulse: $\tau_{\text{on}} = 125$ ps, $\tau_{sw} = 110$ fs. (b) Optical Kerr effect signal generated by the switching laser pulse from (a), focused ($f/30$) into 300 torr, 300 K CO_2 gas. (c) Expanded region from (b) showing rotational wave packet dynamics and field-free alignment in CO_2.

The OKE signal intensity is given by $I(\Delta t) = [\langle \cos^2\theta \rangle - \frac{1}{3} + C]^2$ [15]. The constant C describes the heterodyned signal contribution. Here the polarization rotation by the cell windows provided the local oscillator field for the heterodyne contribution. In Fig. 3(b), we plot the observed signal as a function of time delay between the switching and probe pulses. During their overlap, there is both an electronic and a nuclear component to the signal. After the switching of the 1.064 μm pulse, there is clear rotational revival structure and associated molecular axis alignment indicative of wave packet formation (we note, however, that, due to the room temperature of the gas sample, the macroscopic alignment is small). In this case, the revival structure decays in amplitude due to collisional decoherence. We note that the extension of the switched rotational wave packet method using elliptically polarized light provides a convenient route to field-free three-dimensional molecular alignment [16].

We next consider the limiting case of a switched wave packet in which the interaction with the laser field is dominated by the dipole interaction term, Eq. (3), an example being a coupled quantum well system. These systems respond to the instantaneous electric field and show dynamics dependent upon the carrier phase of the electric field relative to the envelope. Quantum well systems typically exhibit miniband widths much smaller than the interband separation. We therefore neglect the effects of interband transitions, and consider only the intraband dynamics. Within the tight binding (nearest neighbor) approximation, the Hamiltonian for a particle subject to a periodic potential is [17]

$$H_0 = \sum_{n=1}^{N} E_n |n\rangle\langle n| + \sum_{n=1}^{N-1} \Omega_n (|n\rangle\langle n+1| + |n+1\rangle\langle n|), \tag{8}$$

where N is the number of wells. This Hamiltonian is expressed in terms of the Wannier states $|n\rangle$ [18] with on-site energy E_n localized on the well n. Ω_n is the coupling between the n and $n+1$ wells. The interaction with a laser field polarized along the interwell direction is $H_I = -E(t)\mu = -edE(t)\sum_{n=1}^{N} n|n\rangle\langle n|$, where d is the well spacing. If the interaction is turned on adiabatically, each eigenstate of the field-free Hamiltonian will evolve into a single Floquet state. The quasienergy of the kth Floquet state is given by $\epsilon_k(t) = J_0[ed\mathcal{E}(t)/\omega]E_k$ [17]. The degeneracy at the zeros of the Bessel function means that the Wannier states $|n\rangle$ become eigenstates, electron tunneling is coherently suppressed, and dephasing of the Floquet states is inhibited [17]. This fact has previously been explored in relation to creating charge localization in such systems [19]. We show here that a switched wave packet generates time-dependent field-free charge localization. We expand the time-dependent wave function in terms of Wannier states, $\psi(t) = \sum_j a_j(t)|j\rangle$, and propagate numerically the equations of motion of the $a_j(t)$

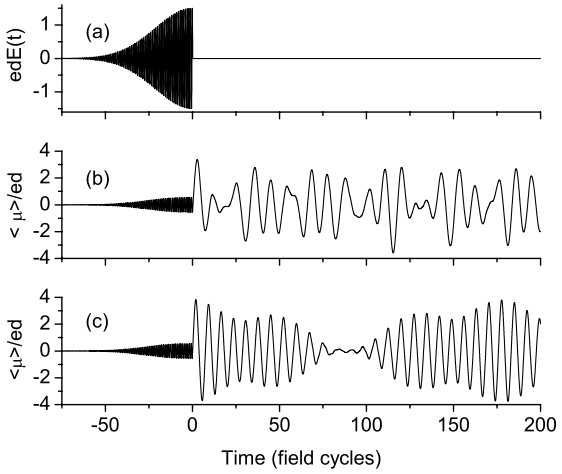

FIG. 4. Switched wave packets in a set of ten coupled quantum wells initially in the ground eigenstate. On-site energies are ϵ_j and $\Omega_j/\omega = 0.25$ for all wells. (a) Laser electric field $ed\mathcal{E}_0/\omega = 1.5$ with $\tau_{\rm on} = 30$ cycles, $\tau_{sw} = 0.1$ cycles. (b) Induced dipole moment for carrier phase $\phi = 0.0$. (c) Induced dipole moment for carrier phase $\phi = \pi/4$.

coefficients. The crossing of the Floquet states restricts the value of $ed\mathcal{E}(t)/\omega$ to $\lesssim 2$ if adiabatic behavior is to be maintained. In Fig. 4, we show the creation of a switched wave packet in an array of ten wells with equal coupling strengths. The laser pulse, Fig. 4(a), has the form

$$E(t) = \mathcal{E}(t)\cos(\omega t + \phi),$$
$$\mathcal{E}(t) = \begin{cases} \mathcal{E}_0 e^{-t^2/\tau_{\rm on}^2}, & t < 0, \\ \mathcal{E}_0 e^{-t^2/\tau_{sw}^2}, & t > 0, \end{cases} \quad (9)$$

where ϕ is the carrier phase of the electric field relative to the envelope. Since the Floquet states are $2\pi/\omega$ periodic, we require $\tau_{sw} \ll 2\pi/\omega$ to maintain the field induced superposition. In Figs. 4(b) and 4(c), we plot the induced dipole moment (charge localization) for two choices of the absolute carrier phase. During $\tau_{\rm on}$, we observe oscillations at the field frequency ω with only moderate localization as the tunneling splitting is reduced by the oscillating field. After τ_{sw}, the superposition evolves under the field-free Hamiltonian and strong time-dependent localization is observed, corresponding to induced charge oscillation in the wells. This localization is substantially larger than during the presence of the laser field and is seen to depend upon the carrier phase. This example is an illustration of a quantum control scenario requiring absolute carrier-envelope phase control [20].

In this Letter, we have explored the use of a strong dynamic Stark interaction with a quantum system for the creation of novel field-free superposition states. The evolution of these field-free switched wave packets contains information about the nature of the strong interaction as well as providing field-free localization. This nonresonant, nonperturbative, but nonionizing regime of interaction could provide new routes to the control of molecular dynamics via the time-dependent modification of an effective Hamiltonian. Control is exerted by reversibly modifying the propagator *during* propagation, as opposed to using only the field-free propagator or the multiple propagators implicit in strong field ionization.

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