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Publisher's version / Version de l'éditeur:
https://doi.org/10.1103/PhysRevLett.92.093001
Physical Review Letters, 92, 9, p. 093001, 2004-03-03

Coherent Control of Rotational Wave-Packet Dynamics via Fractional Revivals

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(Received 5 September 2003; published 3 March 2004)

We show (i) how the evolution of a wave packet created from an initial thermal ensemble can be controlled by manipulating interferences during the wave packet's fractional revivals and (ii) how the wave-packet evolution can be mapped onto the dynamics of a few-state system, where the number of states is determined by the amount of information one wants to track about the wave packet in the phase space. We illustrate our approach by (i) switching off and on field-free molecular axis alignment induced by a strong laser pulse and (ii) converting alignment into field-free orientation, starting with rotationally cold or hot systems.

DOI: 10.1103/PhysRevLett.92.093001 PACS numbers: 33.80.Rv, 33.80.Wz, 42.50.Hz

Once a localized wave packet is excited, its initial localization is quickly lost: the wave packet spreads along the classical orbit which it follows. However, the quantized nature of the spectrum may lead to periodic revivals of the wave-packet localization [1]. At certain fractions of the revival time—fractional revivals [1]—the wave packet relocalizes into several equally spaced [2] copies with well-defined relative phases.

The central idea of this Letter is to control the wave-packet evolution by inducing relative phase shifts between these copies at fractional revivals. Our approach combines aspects of the control methods of Tannor, Kosloff, and Rice [3] and Brumer and Shapiro [4]. Like the former, it uses wave-packet motion. Like the latter, it relies on the interference of multiple pathways—but here these pathways originate from multiple copies of the initial wave packet. Created at fractional revivals, they play the role of "slits" in further evolution.

Dealing with wave-packet dynamics, e.g., in molecules, one is often interested in seemingly simple choices: is a molecular axis aligned or not, where is the vibrational wave-packet localized, etc. At this level one is interested only in the general aspects of wave-packet evolution rather than detailed information such as amplitudes and phases of specific eigenstates. Such limited information is further justified when the wave packet is created from a thermal initial distribution.

We show that, if only a limited amount of information about the wave-packet position or localization is desired, its evolution can be effectively mapped onto a few-state system. The number of "states" is determined by the amount of information one wants to track and/or the degree (detail) of control one wants to exert. The existence of the mapping relies on the presence of revival structures in the wave-packet dynamics. Our basis states are time periodic on the fractional revival time scale, reminiscent of the Floquet states. They track the evolution of the wave-packet symmetries on the spatial and temporal scales of the fractional revivals.

We illustrate our ideas using molecular axis alignment and orientation as examples. We show how relatively weak and short control pulses can implement analogs of $\pi/2$ pulses in the effective few-state system, turning off and on previously induced field-free alignment, and how alignment can be converted into field-free orientation.

Molecular alignment and orientation are long-standing goals, see, e.g., [5,6]. Recently proposed applications include compression of pulses in rotationally excited media [7], control of high harmonic generation [8] or photo absorption and molecular switching [6]. Field-free alignment can be achieved using impulsive [9,10] or switched [11] excitation, with possible improvement using sequences of short pulses [12].

Consider first a 2D rigid rotor confined to a plane (the 3D molecular rotors are discussed later). Its eigenstates are $\psi_M(\theta) = (1/\sqrt{2\pi})e^{iM\theta}$ and the spectrum is $E_M = BM^2$, where B is the rotational constant. A linearly polarized laser field creates an angular potential $U_a(\theta) = -U_0\cos^2\theta$. It aligns the rotor with the direction of laser polarization [6], creating a rotational wave packet $\Psi(\theta, t = 0) = \sum_M a_M \psi_M(\theta)$ localized near $\theta = 0, \pi$. Note that $U_a(\theta)$ is π periodic and does not change the parity of the initial state.

Consider now the free evolution of an initially aligned even state $\Psi(\theta, t = 0) \propto \exp(-\beta\theta^2) + \exp(-\beta(\theta - \pi)^2)$, which contains only even $M \equiv 2n$ states (our following discussion could have equally used only odd states). The spectrum of this subset is $E_n = 4Bn^2$. The "up" and "down" ($\theta = 0$ and $\theta = \pi$) are not distinguished. Shifting θ by π is equivalent to the 2π rotation of the effective angular variable $\tilde{\theta} = 2\theta$: $\psi_n(\theta) \sim e^{i2n\theta} = e^{in\tilde{\theta}}$.

Figure 1(a) shows spreading and revivals of this wave packet $\Psi(t = 0)$ with $\beta = 20.0$ and $B = 1$ [Fig. 1(b)], over the revival period $T_{\rm rev} \equiv 2\pi/(E_n''/2!) = \pi/2B$, in the form of a quantum carpet [13–15]. At $T_{\rm rev}/2$ the initial wave packet revives into a single copy shifted by $\pi/2$, and at $T_{\rm rev}/4$ it splits into two copies spaced by $\pi/2$.

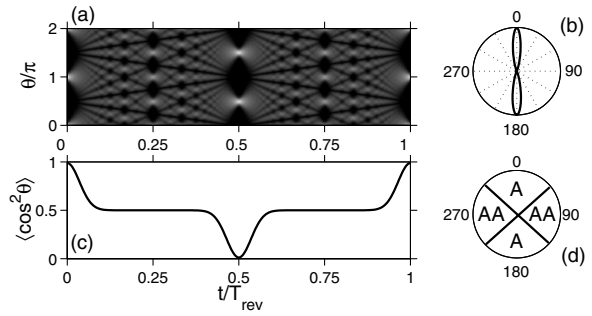

FIG. 1. (a) Evolution of an angular wave packet initially localized at $\theta = 0$ and $\theta = \pi$. Absolute value of the wave function is shown by brightness with black being zero amplitude. (b) Angular distribution for initial wave packet $\Psi(t)$. (c) Time evolution of wave packet's alignment. (d) Aligned (A) and antialigned (AA) regions on the rotor's trajectory.

In general, at $T_{\rm rev}/2K$ the wave packet splits into K copies. These copies are our slits from which the multiple pathways in Fig. 1(a) originate: modifying their phases controls interferences in the subsequent evolution.

Figure 1(c) shows that the commonly used measure of alignment—$\langle\Psi(t)|\cos^2\theta|\Psi(t)\rangle$—provides only limited information about the rich dynamics between the revivals. However, if we are only interested in controlling alignment, this measure is adequate: it is enough to know whether the wave packets are localized in the aligned (A) or antialigned (AA) regions [Fig. 1(d)].

While $|\Psi(t=0)\rangle \equiv |A\rangle$ is maximally localized in the aligned region, $|\Psi(t=T_{\rm rev}/2)\rangle \equiv |AA\rangle$ is maximally localized in the antialigned region and is $|A\rangle$ rotated by $\theta = \pi/2$ ($\tilde{\theta} = \pi$). Their free evolution during $T_{\rm rev}/4$ is

$$e^{-iH_0T_{\rm rev}/4}|A\rangle = \frac{1}{\sqrt{2}}[-e^{i\pi/4}|A\rangle + e^{i\pi/4}|AA\rangle],$$
$$e^{-iH_0T_{\rm rev}/4}|AA\rangle = \frac{1}{\sqrt{2}}[-e^{i\pi/4}|AA\rangle + e^{i\pi/4}|A\rangle]. \quad (1)$$

Superpositions of $|A\rangle$ and $|AA\rangle$ can yield arbitrary alignment between its maximal and minimal values. If we only care about wave-packet localization in A or AA regions, as does the crude measure $\langle\cos^2\theta\rangle$ in Fig. 1(c), we can look at the rotor only at multiples of $T_{\rm rev}/4$. Equations (1) allow us to find eigenstates of the evolution over $T_{\rm rev}/4$:

$$|0\rangle = \frac{1}{\sqrt{2}}[|A\rangle + |AA\rangle]; \qquad |1\rangle = \frac{1}{\sqrt{2}}[|A\rangle - |AA\rangle]. \quad (2)$$

These are analogs of the Floquet states: each revives at every multiple of $T_{\rm rev}/4$, up to a global phase which defines the quasienergy of the state. Using Eq. (1) one can check that the quasienergies are $\epsilon_0 = 0$ for $|0\rangle$ and $\epsilon_1 = 2\pi/T_{\rm rev}$ for $|1\rangle$. Figures 2(a) and 2(b) show the evolution of $|0\rangle$ and $|1\rangle$ for the 2D rotor. They reproduce themselves every $T_{\rm rev}/4$; the dynamics in between remains unnoticed by $\cos^2\theta$.

The field-free alignment revivals can be switched on or off by controlling the relative phase of the aligned and

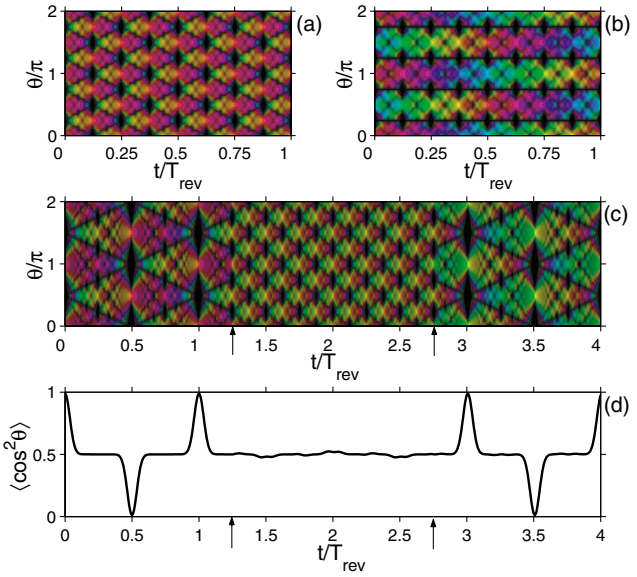

FIG. 2 (color online). (a),(b) Evolution of the states $|0\rangle$ and $|1\rangle$ on the revival time scale. Wave function amplitude is shown by brightness and the phase is shown by shading (color—or hue in the HSL color system). (c),(d) Evolution of the wave function and of $\langle\cos^2\theta\rangle$ for the alignment being switched off and on. Arrows indicate the moments when the laser pulses are applied.

antialigned wave packets at $T_{\rm rev}/4$. Indeed, the field-free evolution turns the initially aligned state $|A\rangle \propto |0\rangle + |1\rangle$ (Fig. 1) into $|0\rangle - i|1\rangle$ at $T_{\rm rev}/4$, with antialigned lobes shifted by $\pi/2$ relative to the aligned ones. Adding $\pi/2$ phase to the aligned lobes transforms the state $(|0\rangle - i|1\rangle)/\sqrt{2}$ to $|0\rangle$ (up to a global phase), switching off alignment revivals. In Figs. 2(c) and 2(d) the instantaneous phase kick $\exp[i(\pi/2)\cos^2\theta]$ was applied at $t = T_{\rm rev} + T_{\rm rev/4}$. The control pulse stops the evolution observed by the crude $\cos^2\theta$ measure [Fig. 2(d)]. Adding the $\pm\pi/2$ phase to these lobes again at $t = 2T_{\rm rev} + 3T_{\rm rev/4}$ changes $|0\rangle$ back to $|0\rangle \pm i|1\rangle$, resurrecting the revivals.

Thus, given the ability to perform phase operations on the wave packets, we could treat and control the rotor dynamics as if we were dealing with only a two-level system. A short linearly polarized pulse acting on a polarizable molecule, which turns $V(\theta, t) = -\frac{1}{4}\alpha_\Delta I(t)\cos^2\theta$ on and off before the wave packets had a chance to move, imparts the phase $\delta\varphi = \exp[-i\int_0^\tau V(\theta, t)dt]$. Here $I(t)$ is the intensity of the control field, $\alpha_\Delta = \alpha_\parallel - \alpha_\perp$, $\alpha_\parallel$ and $\alpha_\perp$ are the parallel and perpendicular polarizabilities, and τ is the pulse duration. $\delta\varphi$ can be used to induce phase shifts between different packets at fractional revivals. Results of finite-time control pulses acting on a thermal ensemble of O_2 molecules are shown below (Fig. 4).

So far we have considered a rotor without the permanent dipole moment, not distinguishing between up and down directions in Fig. 1(d). Consequently, only even $M = 2n$ were used. Consider now an oriented rotor with a permanent dipole μ and spectrum $E(M) = BM^2$ where M is both even and odd. The revival period now is $T_{\rm rev} \equiv$

$2\pi/(E''_M/2!) = 2\pi/B$. Similar to alignment, the evolution can be described on the most coarse-grained scale as a succession of superpositions of oriented $|\text{up}\rangle$ and antioriented $|\text{down}\rangle$ states, with basis states $|0'\rangle = (|\text{up}\rangle + |\text{down}\rangle)/\sqrt{2}$ and $|1'\rangle = (|\text{up}\rangle - |\text{down}\rangle)/\sqrt{2}$. Up to the replacing $\theta \Leftrightarrow \tilde{\theta}$, the quantum carpets for $|0'\rangle$ and $|1'\rangle$ are qualitatively similar to those in Figs. 1 and 2.

At $T_{\text{rev}}/4$ the oriented state $|\text{up}\rangle = (|0'\rangle + |1'\rangle)/\sqrt{2}$ becomes $|0'\rangle - i|1'\rangle$ and is similar to an aligned state. This suggests that aligned state can be converted into the oriented one. If the aligned state is made of even M, it looks like $|0'\rangle$ and is converted into $|0'\rangle - i|1'\rangle$ by a $\pi/2$ relative phase shift of the up lobe. Further free evolution turns it successfully into $|\text{down}\rangle$ (antioriented), $|0'\rangle + i|1'\rangle$ and $|\text{up}\rangle$ (oriented) after $T_{\text{rev}}/4$, $T_{\text{rev}}/2$, and $3T_{\text{rev}}/4$, respectively. The required phase shift can be imparted by a "half-cycle" THz pulse [16] interacting with a permanent dipole, $U(\theta, t) = -\mu E(t)\cos\theta$.

Initial temperature adds a serious complication: for odd M the aligned state is similar to $|1'\rangle$. The same $\pi/2$ relative phase shift of the upper lobe converts it into $|0'\rangle + i|1'\rangle$, yielding $|\text{up}\rangle$ (oriented), $|0'\rangle - i|1'\rangle$ and $|\text{down}\rangle$ (antioriented) after $T_{\text{rev}}/4$, $T_{\text{rev}}/2$, and $3T_{\text{rev}}/4$, respectively. Equal incoherent mixture of $|0'\rangle$ and $|1'\rangle$ (thermal ensemble) yields zero net orientation at these times.

This difficulty is overcome by manipulating the wave-packet interferences at a finer time scale, applying phase shifts at multiples of $T_{\text{rev}}/8$ rather than $T_{\text{rev}}/4$. To describe arbitrary revival pattern at multiples of $T_{\text{rev}}/8$, we must break the orbit $\theta = 0 \ldots 2\pi$ into four parts and look at the superpositions of the four localized states, $|\text{right}\rangle$, $|\text{left}\rangle$, $|\text{up}\rangle$, and $|\text{down}\rangle$. Diagonalizing $\exp[-iH_0 T_{\text{rev}}/8]$ as a 4×4 matrix in the basis of these states, we find four Floquet-like eigenstates which revive every $T_{\text{rev}}/8$ [17]:

$$\begin{aligned} |00\rangle &= (|\text{up}\rangle + |\text{left}\rangle + |\text{down}\rangle + |\text{right}\rangle)/2, \\ |01\rangle &= (|\text{up}\rangle + |\text{left}\rangle - |\text{down}\rangle - |\text{right}\rangle)/2, \\ |10\rangle &= (|\text{up}\rangle - |\text{left}\rangle + |\text{down}\rangle - |\text{right}\rangle)/2, \\ |11\rangle &= (|\text{up}\rangle - |\text{left}\rangle - |\text{down}\rangle + |\text{right}\rangle)/2. \end{aligned} \tag{3}$$

All four orthogonal states are either odd or even on the whole orbit $0 \ldots 2\pi$ and on each of its halves. The state $|00\rangle$ has quasienergy $\epsilon_{00} = 0$, the states $|01\rangle$ and $|11\rangle$ have $\epsilon_{01} = \epsilon_{10} = 2\pi/T_{\text{rev}}$, and $\epsilon_{11} = 8\pi/T_{\text{rev}}$.

The two aligned states are expressed in this basis as $|0'\rangle = (|00\rangle + |10\rangle)/\sqrt{2}$, and $|1'\rangle = (|01\rangle + |11\rangle)/\sqrt{2}$. Their evolution at $T_{\text{rev}}/8$ and $3T_{\text{rev}}/8$ is different: $|0'\rangle$ revives at the equator ($\theta = \pi/2, 3\pi/2$) while $|1'\rangle$ revives at the poles ($\theta = 0, \pi$) [Fig. 3(a) and 3(b)]. The half-cycle pulse $U(\theta, t) = -\mu E(t)\cos\theta$ applied, say, at $3T_{\text{rev}}/8$ imparts the phase shift to the two lobes of $|1'\rangle$ while inducing no relative phase shift to the two lobes of $|0'\rangle$. After $T_{\text{rev}}/4$ the state $|1'\rangle$ becomes oriented while $|0'\rangle$ is again localized at the equator [Fig. 3(a) and 3(b)]. For an equal mixture of even and odd M states in the initial ensemble, half of the ensemble can be oriented while another half of the ensemble remains unoriented and localized at the equator, Fig. 3(c). The pulse also kicks the lobes of the state $|1'\rangle$ at the equator, but the acquired momentum is small compared to the momentum spread there: only small orientation of $|1'\rangle$ is created [Fig. 3(c), thin line].

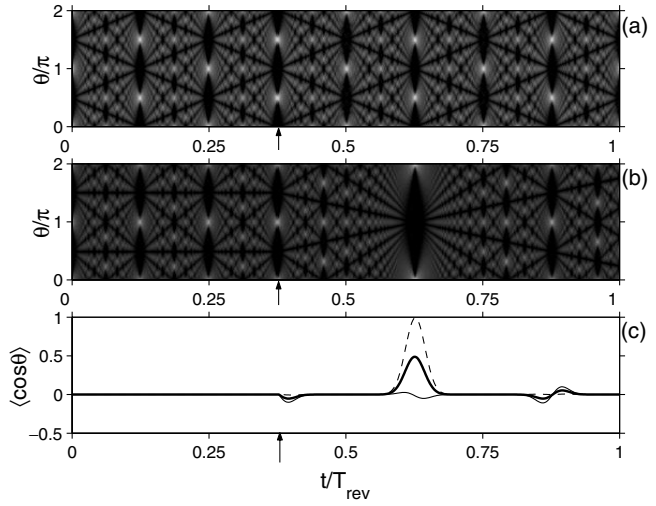

FIG. 3. (a),(b) Evolution of the states $|0'\rangle$ and $|1'\rangle$ in the control scenario discussed in the text. Brightness shows probability density. (c) Orientation of the state $|1'\rangle$ (dashed line), $|0'\rangle$ (thin line), and of their incoherent sum (thick line). Laser pulse is shown by the arrows.

So far we have concentrated on the 2D rotor. The same scheme can be used in 3D, with minor adjustments. First, in 3D $|\text{right}\rangle$ and $|\text{left}\rangle$ overlap due to axial symmetry, but we do not act on them. Second, at $T_{\text{rev}}/8$ the linear term in the 3D spectrum $E(J) = J(J+1)$ shifts revived wave packets by $\pi/4$ from the poles and the equator. The control pulses are applied slightly before or after the exact revival, when the revived wave packets are at the pole and the equator.

Figure 4(a) demonstrates switching off and on rotational revivals for an ensemble of O_2 molecules ($B_0 = 1.4297\ \text{cm}^{-1}$, $D_e = 4.839 \times 10^{-6}\ \text{cm}^{-1}$, and $\alpha_\Delta = 1.099\ \text{Å}^3$ [18]) with temperature $T = 50$ K. The initial alignment is created using a short Gaussian pulse ($I(t) = I_0 \exp[-4\ln 2(t/\tau_p)^2]$) with peak intensity $I_0 = 1.5 \times 10^{14}\ \text{W/cm}^2$ and FWHM duration of the pulse $\tau_p = 50$ fs. This pump pulse was chosen to match that used in recent experiments [10] where the peak intensity was set just below the ionization threshold to ensure strong alignment. The aligning pulse has its peak at $t = 50$ fs in the figure. The revivals are switched off and on by applying a weaker Gaussian control pulse ($I_0 = 0.18 \times 10^{14}\ \text{W/cm}^2$, $\tau_p = 50$ fs) at times 4.300 and 10.150 ps. Note that for the 3D simulations θ is now the 3D polar angle.

Figure 4(b) shows how a half-cycle pulse converts alignment of the carbon sulfide (OCS) molecule ($B = 0.2039\ \text{cm}^{-1}$, $\alpha_\Delta = 4.1\ \text{Å}^3$, $\mu = 0.709$ [19]) into orientation. The initial alignment is created using a Gaussian pulse with $I_0 = 8.26 \times 10^{11}\ \text{W/cm}^2$ and $\tau_p = 50$ fs. The

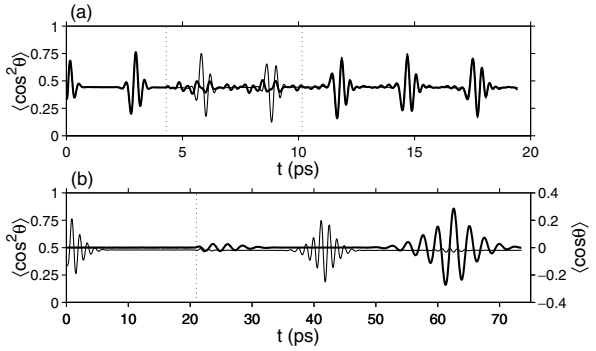

FIG. 4. (a) Alignment of O_2 molecules at 50 K with (thick line) and without (thin line) the control pulses. (b) Alignment (thin line) and orientation (thick line) of OCS molecules at 10 K with the control pulse.

half-cycle pulse used to create orentation ($E(t) = E_0 \exp[-4\ln 2(t/\tau_{hc}^2)]$) has amplitude $E_0 = 8.11 \times 10^5$ V/cm and duration $\tau_{hc} = 500$ fs and was applied 20.536 ps after the aligning pulse.

Unlike in the scheme [20] where the degree of orientation is determined by the half-cycle pulse, here the degree of orientation is determined by the *aligning* pulse. This aspect is similar to Ref. [21] which uses a strong laser field and a weaker dc field to orient a molecule. However, in [21] the strength of the interaction with the dc field μE must amount to kT to get strong orienation, while in our scheme it is the strength of the aligning laser pulse alone that determines the degree of orientation.

Our control scheme is not limited to rotors, but is applicable to any system with quadratic spectrum, with or without the initial temperature, as long as the control interactions depend on the wave-packet position on the orbit. A wave packet centered around n_0 in a system with quadratic spectrum can be viewed as a carrier Ψ_{n_0} modulated by the envelope $f(\theta, t)$ [22], where the angle θ is the phase of the corresponding classical motion. The envelope spreads and revives exactly like a 2D rotor. As for the 2D rotor, the Floquetlike states track the symmetries of the envelope shifted by the linear motion. Their quasienergies and spatial shapes will be discussed in detail elsewhere.

Our scheme gives insight into controlling wave packets for thermal ensembles, when the system starts with an incoherent superposition of many initial eigenstates $n^{(i)}$. Initial laser pulse has to be strong enough to create similar wave packets $\Psi^{(i)}$ for every $n^{(i)}$. If the revival pattern of $\Psi^{(i)}$ is independent of its central state $n_0^{(i)}$ [determined by the initial state $n^{(i)}$], then the control pulses in our scheme would act in a similar way on all wave packets $\Psi^{(i)}$ and need only be concerned with the phase shifts of the wave-packet replicas, unrelated to kT.

Control of thermal ensembles is also possible when the revival dynamics for different $n_0^{(i)}$ is not the same. This is the case with orientation. From the orientation perspective, incoherence of the initial distribution *is not removed* by the aligning pulse. In the envelope description, the wave packet with odd n_0 can be viewed as that with even n_0 (e.g., $n_{0\,\mathrm{even}} = n_{0\,\mathrm{odd}} - 1$), with an additional envelope modulation that turns the even n_0 into odd. An initially incoherent mixture of even and odd states is turned into a mixture of the even ($|0'\rangle$) and the odd ($|1'\rangle$) envelopes for the aligned states. Different revival patterns for $|0'\rangle$ and $|1'\rangle$ allowed us to selectively affect only one of these states while leaving the other virtually unchanged, yielding an incoherent mixture of the oriented state and a state localized near the equator.

We acknowledge fruitful discussions with A. Stolow, J. Underwood, P. Corkum, D. Villeneuve, P. Dooley, and K. Lee, and financial support from NSERC.

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