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**Coherent creation and annihilation of rotational wave packets in incoherent ensembles**Kevin F. Lee,<sup>1,2</sup> E. A. Shapiro,<sup>1,3,4</sup> D. M. Villeneuve,<sup>1</sup> and P. B. Corkum<sup>1,2</sup><sup>1</sup>*Steacie Institute for Molecular Sciences, National Research Council Canada, Ottawa, Ontario, Canada K1A 0R6*<sup>2</sup>*Department of Physics & Astronomy, McMaster University, 1280 Main Street West, Hamilton, Ontario, Canada, L8S 4M1*<sup>3</sup>*Chemistry Department, University of British Columbia, 2036 Main Mall, Vancouver, British Columbia, Canada, V6T 1Z1*<sup>4</sup>*General Physics Institute, Russian Academy of Sciences, 38 Vavilov St. Moscow 117942 Russia*

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Laser pulses can create rotational wave packets in molecules that periodically revive as field-free aligned distributions. These rotational wave packets can be approximately annihilated by applying another laser pulse during a half revival, an effect corresponding to quantum antiresonance in chaotic kicked rotor studies. We theoretically explore causes of deviation from perfect annihilation. We experimentally demonstrate rotational wave packet annihilation in nitrogen gas, measuring the evolution of alignment by Coulomb explosion imaging. As a test, we apply the pulse pair to an existing rotational wave packet and observe the restoration of the original revival structure after the zero-effect pulse pair.

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**I. INTRODUCTION**

The field-free alignment of gas molecules with short laser pulses [1] has recently found many experimental applications. Molecular properties such as refractive index [2–4], ionization rate [5], high harmonic generation efficiency [6,7], and diffraction [8,9] depend strongly on alignment. By controlling rotational wave packets [10–16], alignment-dependent properties are also controlled.

When a short laser pulse illuminates a molecule, the induced dipole will apply a torque. The molecule can then be described as a coherent superposition of rotational states moving towards alignment with the electric field of the laser pulse. Soon after the laser pulse has passed, the rotational wave packet will reach a state of maximum alignment before the system dephases back to a roughly isotropic state. In the rigid rotor approximation, the rotational levels are all related by a fundamental rotational period,  $T_{rev}$ , and thus the wave packet periodically rephases into a state of good alignment [1].

In a recent study, the suppression of alignment was observed by applying a second laser pulse  $T_{rev}/2$  after the aligning pulse [17]. Their analysis found that, for their experimental regime, the alignment signals from individual pulses could be summed, leading to no net alignment [17]. In this paper, we look beyond the alignment signal and ask whether the second laser pulse completely annuls the effect of the first pulse.

If annulment is perfect, then a first laser pulse creates a rotational wave packet from a thermal ensemble, and the second pulse annihilates the wave packet, leaving a thermal ensemble. More generally, the two pulses should form a zero-effect pulse pair, where the two pulses leave no net effect, regardless of the initial state. In this case, an observation would be unable to distinguish whether a zero-effect pulse pair had been applied to the molecules.

To understand the quality of the annulment, we start by analytically evaluating a zero-effect pulse pair. Finding that the annulment is not perfect, we numerically model a wave packet creation and annihilation, and find that the overall

quality of annihilation can still be good. Finally, we experimentally demonstrate annulment by applying a zero-effect pulse pair to a well-defined wave packet, and observing the restoration of the original revival structure after the zero-effect pulse pair.

In terms of molecular control, annihilation can be used to free the timing of the revivals from the fundamental rotational period of the molecule. By annihilating an existing wave packet, a new wave packet can be formed at any later time. Annulment can also be combined with all other multiple-pulse control techniques [12–15], since a series of pulses can be cancelled by starting with the last pulse, and successively annulling the control pulses in reverse order.

Annihilation is a multilevel, multiphoton, rotational analog of a  $0\pi$  pulse in a two-level system. Similar mechanisms have been proposed for vibrational wave packets in  $C_{60}$  [18] and  $H_2$  [19], where vibrations would be stopped by applying a laser-induced dipole force at the right time during an oscillation.

Annihilation appeared in a different form many years ago in the periodically kicked quantum rotor, a staple of quantum chaos studies. When the kicking period is the same as the time between a zero-effect pulse pair, the result is quantum antiresonance [20,21]. When at antiresonance, the system does not gain energy in spite of the numerous kicks. Because adjacent pulses form zero-effect pulse pairs, every other pulse removes the energy added by the preceding pulse. This effect has previously been demonstrated with cold atoms in an optical lattice [22]. As real quantum rotors, rotating molecules could also be used to study quantum chaos [23]. They may offer a more natural way to study decoherence in kicked rotors [24] since collisional decoherence can be controlled by the gas pressure.

The zero-effect pulse pair is related to other rotational wave packet control techniques. Superficially, annihilation appears similar to revival suppression with phase manipulation [15], in that there is no longer net alignment. However, in the phase manipulation case, the wave packet still has the rotational energy gained during alignment, and it still exhibits its higher-order revivals. If the molecules started as a thermal

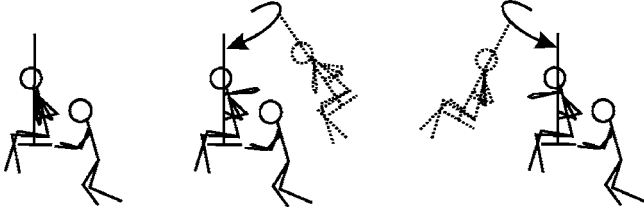


FIG. 1. Illustration of the swing analogy for rotational wave packets. From left to right, pushing the swing represents aligning, enhancing, and annihilating wave packets.

distribution, annihilation returns the wave packet to the same isotropic thermal distribution. In the ideal case, measurements performed after creation and annihilation should be indistinguishable from measurements taken without the pulse pair.

The annihilation mechanism is closely related to enhanced alignment experiments [11–13]. To enhance alignment, a second laser pulse, of intensity comparable to the original aligning pulse and with the same polarization, is applied just before the molecules reach a state of strong alignment. This is analogous to pushing a child's swing at the lowest point in the same direction as the swing is moving, speeding up the swing, as illustrated in Fig. 1. At the middle of the full revival  $T_{rev}$  after the aligning pulse, the wave packet is similar in form to when it was first illuminated by the laser. At this time, a second pulse repeats the action of the original aligning pulse. The average rotational energy increases, and alignment occurs over a shorter time, increasing the peak alignment [12].

To annihilate, we apply the second pulse at the center of a half revival  $T_{rev}/2$  after the aligning pulse, rather than a full revival. At the half revival, the wave packet is also isotropic, but the motion of the molecules is from alignment towards antialignment (perpendicular to the alignment axis [25]). Another laser pulse here is equivalent to hitting the swing opposite to its motion, stopping the swing.

Extending the swing analogy, a swing really corresponds to only one of the rotational states in the wave packet. There are actually many swings involved, so hitting all the swings at the same time, as the laser pulse does to the rotational wave packet, will have the desired effect on all the swings only if they are all in phase during a revival.

## II. THEORY

For a molecule in a linearly polarized laser field of intensity  $I$ , the effective Hamiltonian, averaged over the oscillating field [26,27], is

$$\hat{H} = \hat{H}_0(R, \theta) + V(\theta, t)$$

$$V(\theta, t) = -\frac{I(t)}{2}(\alpha_{\perp}(R) + \Delta\alpha(R)\cos^2 \theta), \quad (1)$$

where  $\hat{H}_0(R, \theta)$  is the field-free Hamiltonian,  $R$  is the internuclear distance,  $\theta$  is the angle between the molecular and laser polarization axes,  $\alpha_{\perp}(R)$  and  $\alpha_{\parallel}(R)$  are perpendicular

and parallel polarizabilities, and  $\Delta\alpha = \alpha_{\parallel} - \alpha_{\perp}$ .

Consider laser pulses of pulsewidth  $\tau$ , where  $\tau$  is much shorter than the average rotational period. Each pulse applies the potential  $V(\theta, t)$  during  $\tau$ . If the field-free motion during  $\tau$  is negligible,  $H_0\tau \ll 1$ , and the propagator  $\hat{U}_{kick}$  is reduced to the coordinate-dependent phase shift

$$\hat{U}_{kick}(\tau) = e^{-i\int_0^{\tau}(\hat{H}_0 + V(\theta, t))dt} \approx e^{-i\int_0^{\tau}V(\theta, t)dt}. \quad (2)$$

For  $V(\theta, t)$  we omit terms that are independent of  $\theta$ , since they only add an overall phase to the whole wave function. For the ac Stark shift [Eq. (1)], this means omitting the  $\alpha_{\perp}$  term. Thus, we have  $-\int_0^{\tau}V(\theta, t)dt = \epsilon\cos^2 \theta$ , where  $\epsilon = \int_0^{\tau}I(t)\Delta\alpha dt/2$ .

### A. Ideal case: 2D rotor

We begin the analysis of annulment in the two-dimensional planar model with only even rotational  $J$  states, a case where annulment is complete. The even  $J$  states do not couple to the odd  $J$  states in this situation. The molecule can only rotate within a plane, and is described by  $\theta$ , the angle of the molecule to the laser polarization. The zero-effect pulse pair (zepp) can then be written, up to an overall phase, as the sequence of three unitary operations  $\hat{U}_{zepp2D}$

$$\hat{U}_{zepp2D} = e^{i\epsilon\cos^2 \theta} \hat{T}_{rev2D/2} e^{i\epsilon\cos^2 \theta}. \quad (3)$$

$\hat{T}_{rev2D/2}$  represents evolution over half of the revival time  $T_{rev2D}$ . Note that we use the more relevant semiclassical definition  $T_{rev2D} = 1/(4B)$  in this two-dimensional section [28], which is half the value of  $T_{rev} = 1/(2B)$  that is used in the rest of this paper for rotations in three dimensions, where  $B$  is the rotational constant of the molecule. For nitrogen,  $T_{rev} = 8.4$  ps [29].

For proper annulment,  $\hat{U}_{zepp2D}$  should be equivalent to  $\hat{T}_{rev2D/2}$ , with the two pulses leaving no net effect. We start by writing out the operators in the basis of the even  $J$  states

$$\cos^2 \theta = \begin{pmatrix} \cdot & \cdot & \cdot & \cdot \\ \cdot & 1/2 & 1/4 & \cdot \\ \cdot & 1/4 & 1/2 & 1/4 \\ \cdot & \cdot & 1/4 & 1/2 \end{pmatrix}, \quad (4)$$

$$\sin^2 \theta = \hat{1} - \cos^2 \theta = \begin{pmatrix} \cdot & \cdot & \cdot & \cdot \\ \cdot & 1/2 & -1/4 & \cdot \\ \cdot & -1/4 & 1/2 & -1/4 \\ \cdot & \cdot & -1/4 & 1/2 \end{pmatrix}, \quad (5)$$

$$\hat{T}_{rev2D/2} = \begin{pmatrix} \cdot & & & \\ & 1 & & \\ & & -1 & \\ & & & \cdot \\ & & & & 1 \\ & & & & & \cdot \end{pmatrix}, \quad (6)$$

with  $\hat{1}$  being the unity operator. Multiplication shows that

$$\hat{T}_{rev2D/2} \cos^2 \theta = \sin^2 \theta \hat{T}_{rev2D/2}. \quad (7)$$

The expanded exponential in  $\hat{U}_{zepp2D}$  is

$$e^{i\epsilon \cos^2 \theta} = \hat{1} + i \cos^2 \theta - \frac{1}{2}(\cos^2 \theta)^2 + \dots \quad (8)$$

Using Eqs. (7) and (8), one can see that

$$\hat{T}_{rev2D/2} e^{i\epsilon \cos^2 \theta} = e^{i\epsilon \sin^2 \theta} \hat{T}_{rev2D/2}. \quad (9)$$

The sequence  $\hat{U}_{zepp2D}$  can then be rewritten as

$$e^{i\epsilon \cos^2 \theta} e^{i\epsilon \sin^2 \theta} \hat{T}_{rev2D/2} = e^{i\epsilon} \hat{T}_{rev2D/2}. \quad (10)$$

That is, up to a global phase, applying  $\hat{U}_{zepp2D}$  to *any* initial state  $\Psi_0$  results in  $\Psi_0$  with  $T_{rev2D}/2$  time evolution, as it would have been if no laser pulses had been applied.

### B. Realistic case: 3D rotor

The 2D, even  $J$  case, represents ideal annulment. We now move to the more realistic model of annulment for a rotor in three dimensions. In 2D, odd  $J$  states were excluded because Eq. (6) does not hold for odd  $J$ , a consequence of the  $BJ^2$  energy spectrum of the planar rotor. In three dimensions, the energy spectrum goes as  $BJ^2 + BJ$ . The extra  $BJ$  term compensates for the different phase accumulation of odd and even states. Both sets of odd and even  $J$  terms will revive in the same way at the half-revival  $T_{rev}/2$ , noting that we have returned to the three-dimensional revival time.

In three dimensions, the sequence of operations for the zero-effect pulse pair becomes  $\hat{U}_{zepp}$

$$\hat{U}_{zepp} = e^{i\epsilon \cos^2 \theta} \hat{T}_{rev/2} e^{i\epsilon \cos^2 \theta}. \quad (11)$$

The matrices for either of the even  $J$  basis or the odd  $J$  basis are given by

$$\cos^2 \theta_{J,J} = \frac{(J+1)^2 - M^2}{(2J+1)(2J+3)} + \frac{J^2 - M^2}{(2J+1)(2J-1)},$$

$$\cos^2 \theta_{J,J+2} = \frac{\sqrt{(J+1)^2 - M^2} \sqrt{(J+2)^2 - M^2}}{(2J+3)\sqrt{(2J+1)(2J+5)}},$$

$$\cos^2 \theta_{J,J-2} = \frac{\sqrt{(J-1)^2 - M^2} \sqrt{J^2 - M^2}}{(2J-1)\sqrt{(2J+1)(2J-3)}},$$

$$\sin^2 \theta = \hat{1} - \cos^2 \theta$$

$$\hat{T}_{rev/2} = \begin{pmatrix} \cdot & & & \\ & 1 & & \\ & & -1 & \\ & & & \cdot \\ & & & & 1 \\ & & & & & \cdot \end{pmatrix}, \quad (12)$$

where  $M$  is the additional projection quantum number, with the field only coupling states having the same  $M$ .

The quality of annulment depends on the equality of  $\hat{T}_{rev/2} \cos^2 \theta$  and  $\sin^2 \theta \hat{T}_{rev/2}$ . For states with  $|M| \ll J$ , and  $J \gg 1$ , the matrix elements approach the two-dimensional values, so annulment should be good in this regime, noting that  $\hat{T}_{rev/2}$  is the same for odd and even  $J$  in three dimensions. If  $J - |M| \ll J$  and  $J \gg 1$ , the  $\cos^2 \theta$  matrix elements become small, so the pair of pulses will have little effect.

Evaluating  $\hat{T}_{rev/2} \cos^2 \theta$  and  $\sin^2 \theta \hat{T}_{rev/2}$  for general  $J, M$  shows that the off-diagonal elements match, but the diagonal elements are not equal, since the diagonal elements of  $\cos^2 \theta$  are not  $1/2$ , as in the two-dimensional case. To understand the effect, one can imagine dividing the first pulse into two subkicks. The first subkick is the  $\cos^2 \theta$  matrix with the diagonal elements artificially set to  $1/2$ . The second subkick is a diagonal matrix that compensates for the artificial diagonal elements of the first subkick.

In such a scenario, the first subkick changes the initial state in a way that can be perfectly annulled. The second subkick modifies the phases of this ideal wave packet, changing the way it revives  $T_{rev}/2$  later. Since the diagonal terms depend on both  $J$  and  $M$ , the diagonal elements of different states will differ from  $1/2$  by different amounts. Thus, applying the full pulse results in a  $J$  and  $M$  dependent chirp relative to what one would have if all the diagonal terms were  $1/2$ . This chirp prevents all the different components from reviving at the right point on the trajectory at  $T_{rev}/2$ , so the quality of annulment will depend on the extent of this chirp.

### C. Numerical simulations

As seen above, this annulment process is not intrinsically perfect. In addition, a real aligning pulse is not completely within the impulsive description of rotational excitation. The finite pulse duration should be comparable to the average rotational period of the molecule for good alignment. The first finite pulse means that the wave packet spends time evolving in a laser field, damaging the latter revival. The second finite kick means that the stopping potential is applied not only during the wave packet revival, but also at times before and after the revival, when phases are not as favorable.

To investigate the importance of deviations from perfect annulment, we numerically simulated the creation and annihilation of a rotational wave packet in a thermal Boltzmann distribution of nitrogen rotating in three dimensions. The calculation used 50 fs full width at half maximum (FWHM)  $\sin^2$  shaped pulses, and included centrifugal distortion of the molecule [25].



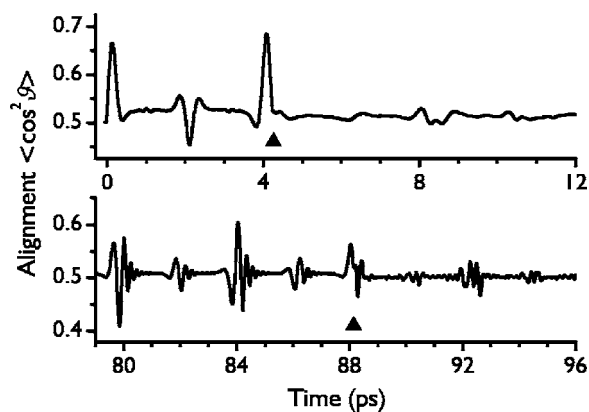


FIG. 2. Calculated revivals in nitrogen. The wave packet is created by a laser pulse at 0 ps. Top: wave packet annihilation at the first half revival at 100 K. Bottom: annihilation at the strongly chirped tenth half revival at 300 K. The triangles indicate the timing of the annihilating pulse.

In the calculation, the creation pulse had a peak intensity of  $7.7 \times 10^{13}$  W/cm<sup>2</sup>. The annihilation pulse, applied 4.21 ps later, most effectively stopped the revivals with a lower peak intensity of  $6.7 \times 10^{13}$  W/cm<sup>2</sup>. The resulting revival structure is shown in the top of Fig. 2 for nitrogen at 100 K. Alignment is measured as  $\langle \cos^2 \vartheta \rangle$ , where  $\vartheta$  is the angle between the polarization axis of the pulse pair, and the projection of the molecule onto a plane containing the polarization axis [25]. A perfectly aligned distribution has a  $\langle \cos^2 \vartheta \rangle$  value of 1, and an isotropic distribution has a  $\langle \cos^2 \vartheta \rangle$  value of 0.5. After annihilation, the revivals are greatly weakened, but there is still some modulation, as seen around 8.4 ps.

To judge the quality of the annihilation, we start with an initial state  $\Psi_0$  that is only in the  $J_0, M_0$  eigenstate. We calculate the state  $\Psi_{final}$  that results from the zero-effect pulse pair, and take the magnitude of the projection  $|\langle \Psi_{final} | \Psi_0 \rangle|$ . For perfect annihilation, the projection would be 1 for all initial eigenstates. Any value lower than 1 indicates population moving to other eigenstates. The resulting projections for the various initial  $J_0, M_0$  states are plotted in Fig. 3. The projections had a temperature-weighted average of 0.867 for 100 K, and 0.912 for 300 K. The projections ranged from 0.57 in the low  $J$  and  $M$  states, to 1.0. The poor recovery of low energy states is likely because they are more easily trapped in the shallower potential before and after the peak of a finite laser pulse. The average projection is better at higher temperatures since there is less population in the lower energy states.

To test the importance of the rigid rotor approximation, we calculated annihilation in 300 K nitrogen at the tenth half revival as shown in the bottom of Fig. 2. These later revivals were noticeably chirped due to centrifugal distortion [25]. The annihilation from the pulse at 88.14 ps was not as clean in this case, but significant revival suppression was still possible, with a weighted average projection of 0.824. Centrifugal distortion changes the frequency of high energy  $J$  states, resulting in poor state projections for certain  $J$  states, depending on the pulse timing. The quality of annihilation in nonrigid rotors thus depends strongly on the degree of popu-

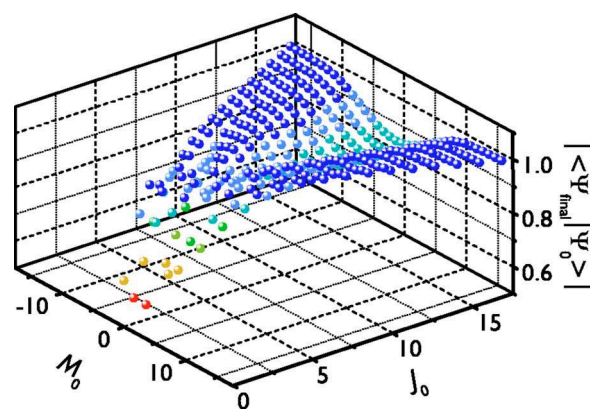


FIG. 3. (Color online) Theoretical projections  $|\langle \Psi_{final} | \Psi_0 \rangle|$  of the wave packet after the zero-pulse pair on the initial rotational eigenstate. A projection value less than 1 indicates undesired population movement to a different eigenstate.

lation of high  $J$  states, which depends on the temperature and the strength of the aligning pulse.

Although not perfect, the important features of annihilation remain in the three-dimensional, finite-pulse calculations. The final wave packet does not exhibit strong alignment, and the wave packet mostly returns to the lower energy rotational states of the original wave function from the higher energy states of the aligned wave packet.

### III. EXPERIMENT

To experimentally demonstrate wave packet annihilation, we create a rotational wave packet with one laser pulse and optimize a second pulse to annihilate the wave packet in a molecular beam of nitrogen in a Coulomb explosion imaging chamber [25]. To verify that this is a zero-effect pulse pair, we apply the same pulse pair to an existing rotational wave packet, rather than a thermal distribution, and observe the return of the original wave packet after the pulse pair. A regenerative amplifier provided 50 fs FWHM, 815 nm laser pulses. The molecular beam of nitrogen had an estimated rotational temperature of about 100 K.

We diagnose alignment at different time delays by multiphoton ionizing the molecules with a  $10^{16}$  W/cm<sup>2</sup> circularly polarized probe pulse. The unbound, positively charged nitrogen fragments rapidly separate along the internuclear axis in a Coulomb explosion. A constant electric field directs the ions towards a microchannel plate backed by a delay-line anode. This detector yields time and position of impact, which can be converted into the fragment velocities at the time of explosion, and thus the alignment of the molecules. The angular distribution for a particular time delay is made by observing a few thousand fragments [25].

The revival structures measured in the experiment are shown in Fig. 4 with alignment plotted as a function of time. As in the calculation,  $\vartheta$  is the angle between the polarization of the linear aligning pulse and the projection of the molecular axis onto the plane of polarization of the circularly polarized probe pulse. This angle is free from the bias inherent in Coulomb explosion with 50 fs pulses [25].

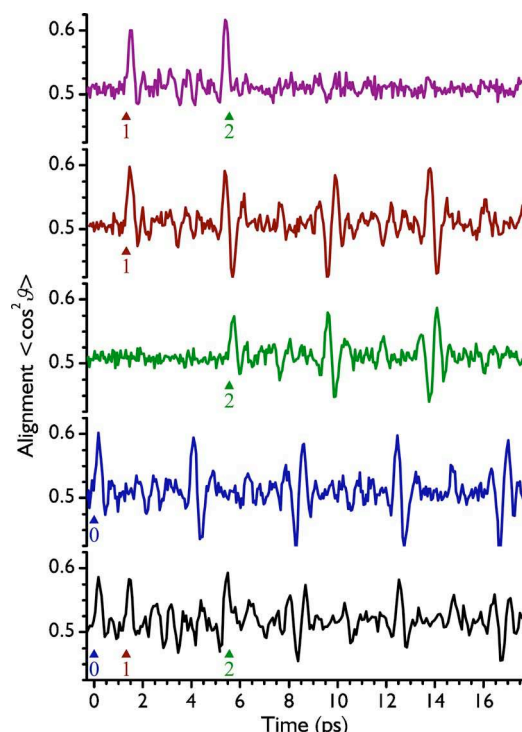


FIG. 4. (Color online) Experimental wave packet annihilation in nitrogen. These are the revival structures from various combinations of the three laser pulses. The numbered triangles indicate which pulses were applied, and their timing. In the top curve, pulse 1 creates a rotational wave packet, which is annihilated by pulse 2. In the lowest curve, pulse 0 creates a test wave packet which is distorted by pulse 1 until pulse 2 annuls pulse 1, restoring the revivals from pulse 0.

Each pulse from the laser was split and recombined to yield four copropagating laser pulses with independent timing, intensity, and polarization. One pulse was the probe pulse, which measured the alignment of the molecules. The other three pulses were used for molecular alignment, and had the same linear polarization. The revival structures resulting from various combinations of the three aligning pulses are shown in Fig. 4.

The three middle curves of Fig. 4 show the revivals from each pulse alone. Pulses 1 and 2 were the zero-effect pulse pair. The timing and intensity of pulse 2 was chosen to minimize the alignment after pulses 1 and 2, shown in the upper

curve. The revival structure with all three alignment pulses is shown in the lowest curve of Fig. 4. Pulse 0 creates a test packet at 0 ps. At 1.3 ps, pulse 1 disrupts the test packet, which no longer revives strongly  $T_{rev}/2$  (4.2 ps) after pulse 0. At 5.6 ps, pulse 2 annuls the effect of pulse 1, restoring the revivals at fractional multiples of  $T_{rev}$  after pulse 0. The wave packet is approximately returned to the state it would have been in if pulses 1 and 2 had not been applied.

Pulses 0 and 1 had peak intensities of about  $3.0 \times 10^{13}$  W/cm<sup>2</sup>. Pulse 1 was placed 1.33 ps after pulse 0, where it clearly disrupted the revivals from pulse 0. Pulse 2 was 4.23 ps (about  $T_{rev}/2$ ) after pulse 1, with a peak intensity of about  $2.7 \times 10^{13}$  W/cm<sup>2</sup>. The relative pulse timing may have a systematic error of up to the 50 fs laser pulse duration. The intensities were roughly calibrated by measuring nitrogen ionization rates at different pulse energies.

#### IV. OUTLOOK

We have shown that the rotational effects of an aligning pulse can be approximately annulled by a second pulse applied half of the fundamental rotational period later. With the observation of wave packet annihilation [17] and enhancement [12,13], quantum chaos studies should be possible with rotational wave packets.

The ability to create and annihilate wave packets at arbitrary times with femtosecond precision, combined with existing multiple-pulse techniques [12–15], may be useful for applications of rotating molecules such as ultrafast refractive index modulation [3,4]. Repeatedly forming new wave packets may improve revival quality at long time scales. Zero-effect pulse pairs might be useful for handling mixtures of different species, isomers, or conformers. If the components have different revival times, a laser pulse at a particular half revival could selectively annihilate the wave packet for only one component.

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