

Phase Control of Rotational Wave Packets and Quantum Information

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Lasers can create rotational wave packets in gas-phase molecules which periodically revive as field-free, aligned distributions. We control the wave packet evolution with relatively weak laser pulses at fractional revivals which modify the phase between wave packet components. We demonstrate two phase control effects in oxygen: coherently switching revivals off and on, and doubling the revival frequency. When viewed as a quantum logic system, these effects correspond to a Hadamard and a T operation.

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Thermal gas-phase molecules point randomly, making it difficult to experimentally probe directional properties of isolated molecules. To overcome this problem, short laser pulses have been used to momentarily align gas molecules, forming rotational wave packets which periodically revive as strongly aligned distributions without the presence of an external field [1–3].

Rotational wave packets have recently been used to measure the dependence of high harmonic generation [4,5] and strong-field ionization [6] on molecular alignment. Using phase modulation from rotational wave packets, 400 nm laser pulses have been compressed below 10 fs [7]. There are proposals to use rotational wave packets for attosecond pulse shaping [4,5], measuring molecular structure with laser-induced electron diffraction [8,9] and molecular orbital tomography [10].

If rotational wave packets can be controlled, then all of the above processes can be controlled. Wave packet evolution is governed by the changing phases of its components. One way to control rotational wave packets is to alter the phases between the components with relatively weak laser pulses applied at particular fractional revival times [11]. This approach is conceptually different from using strong pulses to reshape the wave packet [12,13].

We experimentally demonstrate rotational wave packet control by phase manipulation. Phase manipulation can create any degree of alignment at the appropriate fractional revival time up to the limit set by the number of coherently populated rotational states [14]. We demonstrate the fundamental manipulations that form the basis for this arbitrary wave packet control. These basic manipulations correspond to the ability to repeatedly and coherently destroy and restore the periodic recurrences of net alignment, and to the doubling of the frequency at which alignment occurs.

We adopt the vibrational wave packet quantum logic formalism [15] as a convenient way to describe rotational revival evolution and manipulations. In this form, our basic manipulations correspond to Hadamard, T, and

$\pi/4$ phase operations which can be combined to perform all single-qubit operations [16]. Information processing is another possible application of rotational wave packet phase manipulations.

We modify rotational wave packets created using standard impulsive alignment techniques [17]. In a typical single-pulse alignment experiment, an intense, nonresonant, linearly polarized laser pulse induces a dipole in linear molecules, resulting in a torque that turns the molecules towards the alignment axis as defined by the laser polarization. The pulse passes before the molecules reach peak alignment, after which they rotate freely. Because of rotational J state quantization, the aligned state will recur at multiples and fractions of the fundamental rotation time T_{rev} , the period of the fundamental J state [3,18], which is 11.5 ps for oxygen. Note that there is also a second convention for T_{rev} [11,15,19], which is half of our T_{rev} value. The wave packet evolution is often measured by changes in birefringence [1] or Coulomb explosion imaging [17].

The time evolution of a typical rotational wave packet in oxygen is shown in the upper panel of Fig. 1. The angular distributions measured by Coulomb explosion imaging were converted to the average value $\langle \cos^2 \vartheta \rangle$ as a measure of net alignment. ϑ is the angle between the alignment axis and the molecule's projection onto the measurement plane which includes the alignment axis [17]. The $\langle \cos^2 \vartheta \rangle$ value corresponding to perfect alignment is 1, isotropic is 0.5, and perfect antialignment, with all molecules perpendicular to the alignment axis, is 0. The origin of the time axis is the peak of the aligning pulse. Since oxygen has only odd J states due to its nuclear spin, there are revivals in net alignment at multiples of $T_{\text{rev}}/4$.

Not visible using $\langle \cos^2 \vartheta \rangle$ are the fractional revivals at odd multiples of $T_{\text{rev}}/8$. These are revivals with equal aligned and antialigned components which we call a cross, after its projected shape on the measurement plane. It is at these crosses where we can modify relative phase.

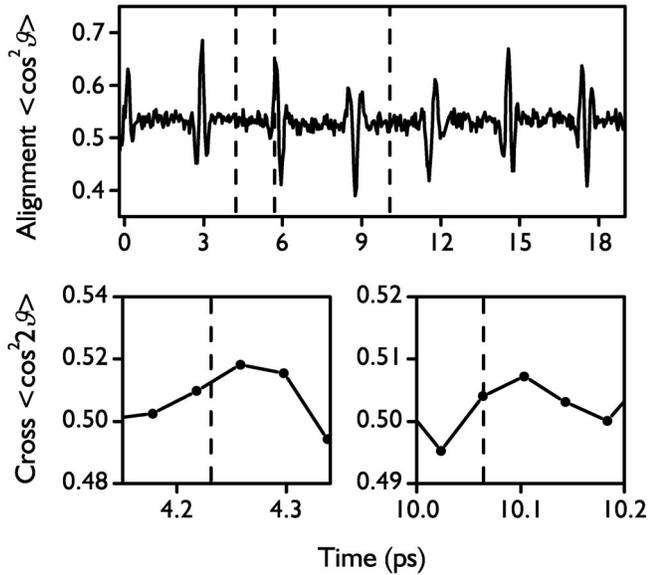


FIG. 1. The upper panel shows the measured time evolution for an unmodified rotational wave packet in oxygen. The lower panels show second order alignment as seen by the modifying pulses. The dashed lines indicate the approximate location in time of the peak of the 50 fs FWHM pulses that performed the phase modifications.

The cross revivals can be seen using $\langle \cos^2 2\vartheta \rangle$, which is 1 for a perfect cross.

The wave packet is conveniently described by separating the cross into an aligned and an antialigned component, $|A\rangle$ and $|AA\rangle$. $|A\rangle$ ($|AA\rangle$) has the same shape as the wave packet when it is aligned (antialigned) at a revival. In the envelope representation [15,20], or the 2D rigid rotor model [11], $|A\rangle$ and $|AA\rangle$ are exact copies of the original wave packet at opposite points of the trajectory with a well-defined relative phase.

The two cross-shaped superpositions

$$|0\rangle = (|A\rangle + |AA\rangle)/\sqrt{2}, \quad (1)$$

$$|1\rangle = (|A\rangle - |AA\rangle)/\sqrt{2}, \quad (2)$$

are eigenstates of discrete time evolution that, in the 2D rotor case, revive exactly every $T_{\text{rev}}/8$ up to a global phase driven by their quasienergy: $\epsilon_0 = 0$, $\epsilon_1 = 4\pi/T_{\text{rev}}$ [11]. In the 3D case, the revival and rotational frequencies do not have an integer ratio. Because of this, the cross shape, where a control pulse would be applied, occurs near, but not exactly at, $T_{\text{rev}}/8$.

Revivals can be seen as a beating of the superposition state, with $|A\rangle = (|0\rangle + |1\rangle)/\sqrt{2}$. Alignment at revivals can be manipulated by controlling the states $|0\rangle$ and $|1\rangle$. It can be convenient to consider $|0\rangle$ and $|1\rangle$ as the two levels of a qubit to make use of quantum logic formalism for creating different alignment states. Inversely, one might consider manipulation of alignment to be operations on a

qubit that can be read by measuring whether or not the wave packet is aligned, antialigned, or in a superposition or mix of these two states.

To control the states $|0\rangle$ and $|1\rangle$, we take advantage of the angular separation of the $|A\rangle$ and $|AA\rangle$ components at a cross revival. When a short laser pulse, polarized along the alignment axis, is applied to a cross, the aligned component will experience a lower potential than the antialigned component, due to the different polarizability of molecules aligned parallel and perpendicular to the laser field. This angular dependence results in a change of relative phase between $|A\rangle$ and $|AA\rangle$. Higher laser intensity will increase the interaction strength and the accumulated phase difference. $T_{\text{rev}}/8$ later, the wave packet will revive into a different superposition of $|A\rangle$ and $|AA\rangle$, depending on the amount of phase added.

By defining the eigenstates in terms of the wave packet itself, we gain robustness against temperature. As long as the molecule can be aligned well, revivals and phase kicks will work in the same way regardless of the initial state of the molecule.

An unmodified rotational wave packet at a cross revival $T_{\text{rev}}/8$ after alignment is written as $(|0\rangle + i|1\rangle)/\sqrt{2} = e^{i\pi/4}(|A\rangle - i|AA\rangle)/\sqrt{2}$. This state can be turned into $|0\rangle$ or $|1\rangle$ by a $\pi/2$ phase change between $|A\rangle$ and $|AA\rangle$. These states always revive as crosses, and thus never exhibit net alignment. By adding $\pi/2$ at a later cross, one can return to a superposition, restoring the net alignment revivals.

We experimentally implement the above two phase effects. Much of our apparatus has been described previously [17]. We use 810 nm, 50 fs FWHM pulses from a regenerative amplifier. Beam splitters split the original pulse into four pulses that are modified in intensity and polarization, then recombined. Translation stages control the relative timing between pulses. A telescope is used to make the probe beam half the size of the other beams at the focus.

The first pulse in time aligns the molecules with 1×10^{14} W/cm². The next two pulses, at about 10^{13} W/cm², apply phase changes. The fourth pulse probes the alignment with 10^{16} W/cm². The four pulses are aligned by optimizing the interference fringes seen between the beams. We have verified this technique using a lens and pinhole outside of the vacuum chamber. We also characterize the externally focused beam to measure the effect of aberration on the focal spot size. The intensity inside the chamber was measured by finding the saturation intensity for ionization of a noble gas and scaling the result to the different beams. We roughly estimate a total error of 20% for the intensity of the beams at the location of the probe focus.

We align oxygen from a skimmed molecular beam with an estimated rotational temperature of 100 K. We chose oxygen for three reasons. The moment of inertia of oxygen is compatible with alignment by 50 fs pulses.

Diatomics are well suited to Coulomb explosion imaging. Oxygen has only odd rotational states which all revive as crosses at the same time, unlike molecules with mixtures of odd and even rotational states which would require a more complex alignment technique [21].

Alignment is measured with a circularly polarized probe pulse that removes about five electrons from oxygen, breaking the bond. The charged atoms repel and separate in a Coulomb explosion. By measuring the final position and time of the fragments in a constant electric field spectrometer, we can calculate the momentum of the fragments when they exploded. For a diatomic, the momentum vector lies along the internuclear axis, so we can measure an angular distribution by measuring the momenta of a few thousand fragments.

To turn alignment off, we first measured the timing of the cross revival as seen in the lower left panel of Fig. 1 to roughly determine the timing of the second pulse. The amplitude of a later revival was minimized using the timing and intensity of the second pulse. To turn the alignment back on, we use the cross at 10 ps, shown in Fig. 1, where the third pulse was optimized in time and intensity by observing the turn-on. In the experiment, the pulse intensities were 2.0×10^{13} W/cm² and 2.3×10^{13} W/cm² for turn-off and turn-on, respectively. In our rotational wave packet simulations, optimal turn-off and turn-on were achieved with 1.9×10^{13} W/cm² with 50 fs pulses.

With both control pulses in place, the evolution was measured as shown in the solid curve of Fig. 2. The dotted reference curve shows conventional alignment with arbitrary vertical scale. Times between the control pulses at

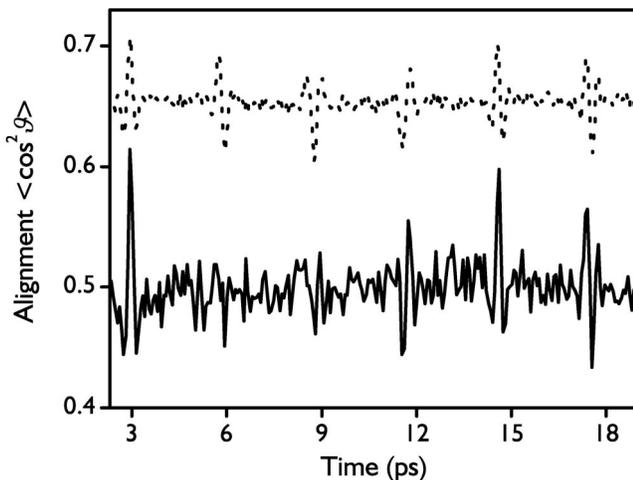


FIG. 2. The solid curve shows the coherent switching off and on of revivals in net alignment from $\pi/2$ phase changes applied to cross revivals at 4.23 and 10.1 ps after the aligning pulse as measured in oxygen. For comparison, the dotted line shows an unmodified wave packet, at arbitrary vertical scale, measured under similar conditions.

4.23 and 10.1 ps do not show significant net alignment. The wave packet is still coherent, as seen by the cross revival shown in the lower right panel of Fig. 1 where the turn-on pulse is applied. Net alignment returns after the restoring pulse at 10 ps.

We now use quantum logic to show how phase control can be used as the basis for complete single-qubit operations on the levels $|0\rangle$ and $|1\rangle$. Viewed this way, $T_{\text{rev}}/8$ time evolution is a $\pi/4$ phase gate since it causes a $\pi/2$ phase shift or, equivalently, opposite $\pi/4$ phase shifts to each eigenstate. Switching alignment on and off corresponds to Hadamard operations, since the system is being brought from the aligned superposition $|0\rangle + |1\rangle$ to an unaligned eigenstate, and back. The full Hadamard gate, which performs a Hadamard operation for any superposition state, can be applied with a similar three-pulse sequence that will be described in a future paper [20].

The natural complement to the Hadamard gate is the $\pi/8$ phase gate, known as the T gate, which adds opposite $\pi/8$ phase shifts to each of the basis states, for a total $\pi/4$ relative phase. If these two gates were available, then any desired superposition of the basis states, or equivalently $|A\rangle$ and $|AA\rangle$, could be created [16].

We demonstrate a T operation by using two pulses to put the original aligning wave packet into a state with a $\pi/4$ phase difference every $T_{\text{rev}}/8$, such as $(|0\rangle + e^{-i\pi/4}|1\rangle)/\sqrt{2}$, as one would get from a T gate. Since time evolution adds $\pi/2$ phase between $|0\rangle$ and $|1\rangle$ every $T_{\text{rev}}/8$, the phase between $|0\rangle$ and $|1\rangle$ always differs by an odd multiple of $\pi/4$. These states all have the shape of an uneven cross which results in net alignment. This leads to the interesting effect of doubling the revival frequency,

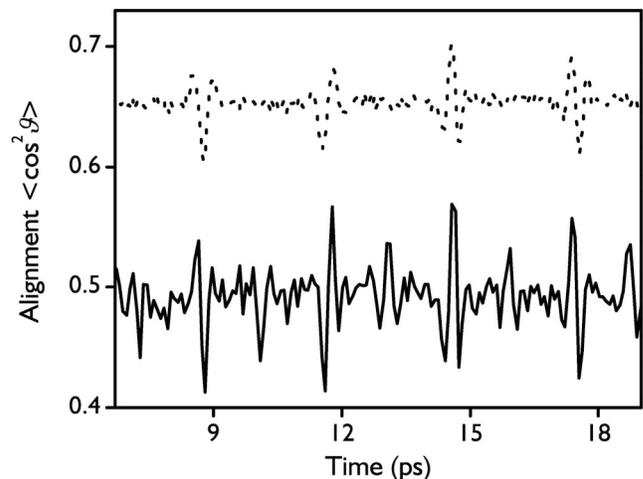


FIG. 3. The solid curve shows the measurement of a doubled revival frequency in oxygen. Conventional revivals, shown by the dotted curve at arbitrary scale, were modified by the addition of $\pi/4$ of phase at the cross at 4.23 ps, and $\pi/2$ at the revival 5.7 ps after the aligning pulse.

since there will be net alignment every $T_{\text{rev}}/8$ instead of $T_{\text{rev}}/4$.

The first pulse in our T operation adds $\pi/4$ of phase to a cross. $T_{\text{rev}}/8$ later, the wave packet has the correct $|A\rangle$ to $|AA\rangle$ ratio of the uneven cross, but the wrong phase. To correct the phase, the second pulse adds $\pi/2$ to this uneven cross, finishing the operation. The full T gate can be performed using a similar three-pulse sequence [20].

We doubled the revival frequency in oxygen, first applying a control pulse at 4.23 ps. This was the same time as for the revival turn-off, but about half as intense to add $\pi/4$ of phase instead of $\pi/2$. The second control pulse, applied at 5.7 ps, was optimized in timing and intensity by the appearance of extra revivals. The pulses had intensities of 0.9×10^{13} W/cm² and 2.1×10^{13} W/cm². In our simulations, optimal pulse intensities were 1.1×10^{13} W/cm² and 1.9×10^{13} W/cm². The measured doubled revivals are shown as the solid curve in Fig. 3, while the dotted curve is an arbitrarily scaled, single-pulse reference. New times of alignment appear between the usual revivals.

We have implemented two operations that are relevant to quantum computing. While we do not claim a complete quantum computing system, there are advantages to revival encoding that make it worth considering. Wave packets can be initialized and manipulated at high temperature. For nonpolar molecules, the radiative lifetime is on the scale of years. Coherence is lost through external interactions which can be partly controlled. Collisions limit coherence to the nanosecond time scale at atmospheric pressure, and much longer at lower pressures. At lower gas densities, operation time is limited by the molecule leaving the laser interaction region.

Two-qubit operations can be performed on multiple qubits encoded in a single rotational wave packet [11,20], but this method has limited scalability. Entangling qubits in many separate molecules is scalable, but requires external coupling. Molecules with permanent or laser-induced dipoles in a trap are one possibility [22], or arrays of surface-attached molecular rotors might serve as qubits, with a rotor on the tip of a scanning positioning microscope as a data bus [23–25], although the result of such a coupling is an open question.

In conclusion, we have demonstrated phase control of rotational revivals in oxygen. Besides applications to quantum information, this is a general technique which can control the external interactions [4–10] of any anharmonic system that exhibits revivals, and has a coordinate-dependent light interaction. This includes vibrating [15] as well as rotating systems.

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