LETTER TO THE EDITOR

Two-pulse alignment of molecules

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Abstract
Field-free alignment of gas-phase molecules can be achieved by creating rotational wavepackets with a short laser pulse. We demonstrate that the degree of alignment can be improved by illuminating the molecules with a second laser pulse at a specific time during the evolution of the original rotational wavepacket.

There has long been an interest in aligning gas-phase molecules [1–3], and now, field-free alignment of molecules using rotational wavepackets [4–8] is quickly becoming an important tool for molecular physics. Molecular alignment has been used to measure the angular dependence of ionization probabilities [9], and high harmonic generation [10, 11]. Field-free alignment can even be used to prepare gas molecules for diffraction experiments [12].

The growing popularity of alignment with rotational wavepackets is due to the quality of field-free alignment that can be achieved by using a single laser pulse. Through interactions with the anisotropic polarizability of a molecule, a laser pulse can induce rotations of the molecule. When applying a laser pulse, the most polarizable axis of the molecule will be torqued into the laser polarization direction. With a sufficiently short pulse duration, the laser pulse will have passed before the molecules reach their peak alignment. The molecules then rotate freely, with regular recurrences of the original aligned distribution due to the nature of the rotational states [1].

There have recently been proposals to modify the normal evolution of rotational wavepackets by applying additional laser pulses to the rotating molecules at selected times after the first aligning pulse [13–16]. One predicted result was that these extra pulses could be designed to successively increase the peak alignment beyond the level achievable by a single aligning pulse [13, 14]. In what follows, we implement this idea by using two pulses to increase the degree of alignment.

We show that a sequence of two laser pulses can be used to generate stronger alignment than is possible for either of the two pulses individually. More generally, we show that a second aligning pulse alters the temporal shape of the rotational revivals. The new revivals
show increased or decreased alignment, depending on the relative timing of the two laser pulses.

We build on our previous work in which we created rotational wavepackets using a single aligning pulse, and measured their evolution with a subsequent probe pulse [7]. To implement and measure two-pulse alignment, we need three laser pulses. The first aligning pulse prepares a rotational wavepacket at time $t_1$ and the second aligning pulse modifies this rotational wavepacket at time $t_2$. At time $t$, a probe pulse is used to measure the resulting alignment.

Our aligning pulses are linearly polarized, so the polarization defines the axis towards which molecules are torqued (alignment axis). We use a circularly polarized probe pulse since it does not exert a torque within the polarization plane. Molecules will still be pulled towards the circular polarization plane. We measure the angle $\vartheta$ between the projection of the molecule on the polarization plane and the alignment axis. Unlike the angle directly between the molecule and the alignment axis, the angle $\vartheta$ will not be changed by a circularly polarized pulse [7].

The circular probe pulse is intense enough to multiply ionize the molecules until they are no longer bound, and the positive ions fly apart in a Coulomb explosion. The resulting molecular fragments are accelerated by a uniform electric field onto a microchannel plate stack backed by a delay-line anode. This detector measures the time and position of impact of each fragment, which can be converted into the initial velocity vector of the fragment immediately after explosion. For diatomics, the velocity vector will point along the direction of the internuclear axis at the time of explosion.

We measure the angular distributions of about 2000 molecules, and calculate the value $\langle \cos^2 \vartheta \rangle$ as a simple way to represent the quality of the alignment. A higher value of $\langle \cos^2 \vartheta \rangle$ corresponds to more molecules pointing along the alignment axis. A $\langle \cos^2 \vartheta \rangle$ value of 1 indicates complete alignment, an isotropic distribution has a $\langle \cos^2 \vartheta \rangle$ value of 0.5 and 0 represents complete antialignment with all molecules perpendicular to the alignment axis.

Our experiments are performed on molecules in a gas jet that we illuminate with 50 fs laser pulses from a titanium:sapphire-based regenerative amplifier. In the nitrogen data that will be presented, the gas jet is formed by expanding 600 Torr through a 25 $\mu$m aperture, then skimming to a roughly 70 $\mu$m width. The linear aligning pulses are attenuated to about $10^{14}$ W cm$^{-2}$ and the circular probe pulse is almost $10^{16}$ W cm$^{-2}$. By using the probe to measure the angular distribution at different times, we can reconstruct the evolution of the rotational wavepacket [7].

A single pulse rotational revival structure is shown in figure 1 ($\sim 1.6 \times 10^{14}$ W cm$^{-2}$, 50 fs). The aligning pulse is applied at time $t_1$ and the probe pulse measures the angular distributions at different times $(t - t_1)$ after the aligning pulse. The molecules are strongly aligned immediately after the aligning pulse, and at half-integer multiples of the fundamental rotational period of 8.5 ps.

When aligning molecules with a single pulse, there are two basic laser parameters, the pulse duration and the pulse intensity. The limits on these parameters are set by properties of the molecule being aligned. Since the aligning intensity determines the strength of the interaction with the molecule, an upper bound on the intensity can limit the degree of alignment achieved. Nonlinear ionization [17] imposes a maximum intensity; however, alignment can still be improved by applying a second pulse, also up to the single pulse limit, without much increase in the ionization rate [13, 14].

When two aligning pulses are used, there are five available parameters: two pulse durations, two intensities and the time delay between the two pulses. There is much space to explore and many ways to play with the rotational wavepackets [13, 15, 16]. In the following
Figure 1. This plot shows the first two full rotational revivals in nitrogen induced by a single pulse to illustrate the pulse timing. The first pulse initiates a rotational wavepacket at time $t_1$. The first full rotational revival occurs near 8.5 ps. In the two-pulse nitrogen experiment, the second pulse was scanned through the region $\|$ around 8.5 ps. For each timing of the second pulse, the resulting two-pulse revival in the 17 ps region $\|/\|$ is measured. Higher values of $\langle \cos^2 \vartheta \rangle$ indicate stronger alignment as depicted by the insets which illustrate a molecule aligned and antialigned to the alignment axis.

In the experiment, we apply the second aligning pulse at eight different times $(t_2 - t_1)$ during the first full revival of the rotational wavepacket, around 8.5 ps (region $\|\|$ in figure 1). For each time delay $(t_2 - t_1)$, we measure the wavepacket revival $\sim 17$ ps after the first aligning pulse (region $\|/\|$ in figure 1).

Two-pulse alignment data for nitrogen are shown in figure 2. The surface is constructed by plotting $\langle \cos^2 \vartheta \rangle$ as a function of the aligning pulse delay $(t_2 - t_1)$, and the probe time $(t - t_1)$. Each set of revivals differs by the location in time of the second aligning pulse $(t_2 - t_1)$, which came from 8.31 to 8.53 ps after the first pulse. As can be seen by the peak in figure 2, alignment was only enhanced for certain time delays between the two aligning pulses, while other time delays will actually decrease the alignment. Different aspects of figure 2 are highlighted in figure 3.

Figure 3(a) shows the changes in the maximum and minimum of the 17 ps revival as the timing of the second pulse is changed. The extent of the 17 ps revival is plotted as a vertical line at the time corresponding to the delay between the two aligning pulses. The curve marked (1) is the revival when only the first aligning pulse is applied, and is plotted to show the state of the rotational wavepacket when the second pulse is applied. The alignment was most strongly enhanced when the second pulse arrived while the molecules were starting to approach high alignment. This was the case for all our measurements, as predicted in [14]. That the optimum time occurs between peaks suggests that the enhancement is achieved in part by increasing the overall rotational energy, since molecules that are neither aligned nor antialigned experience the most torque from the laser pulse [3]. In the case illustrated in figures 2 and 3, the two aligning pulses had similar intensities of roughly $1.3 \times 10^{14}$ W cm$^{-2}$, and the optimal pulse delay was 8.42 ps.
Figure 2. This surface map shows the measured alignment as a function of time $t - t_1$ since the first pulse, and the delay between the first and second aligning pulse, $t_2 - t_1$. Both pulses had similar estimated intensities of $1.3 \times 10^{14}$ W cm$^{-2}$. The dashed curves on the walls are single pulse references. The left curve is the alignment that would occur with the second pulse only. The right curve is the alignment that would occur with the first pulse only.

Figure 3. These plots show the data from figure 2 from different perspectives. Plot (a) shows the effect of the relative pulse timing. The vertical lines indicate the amplitudes of the measured revivals at 17 ps plotted against the pulse delay $t_2 - t_1$. Curve (1) is the full revival that would occur due to the first pulse alone. Alignment is enhanced when the second pulse is applied as the molecules are in the process of realigning. Plot (b) shows the relative improvement in the peak alignment. Curve (2) is the revival that would occur from applying the second pulse only at 8.32 ps. Curve $\circ$ is the two-pulse revival with $(t_2 - t_1) = 8.42$ ps, and curve $\triangle$ has $(t_2 - t_1) = 8.31$ ps.

The curves in figure 3(b) show the two-pulse revivals which had the strongest alignment $(t_2 - t_1 = 8.42$ ps, $\circ)$ and antialignment $(t_2 - t_1 = 8.31$ ps, $\triangle)$. The curve marked (2) shows the revival that was induced by applying only the second pulse at 8.32 ps after the first pulse would have passed. The corresponding revival from only the first pulse was
These data illustrate a strong enhancement of alignment in oxygen using two pulses measured at time $t$ after the first pulse. The curve marked (1) shows alignment resulting from the first pulse only. The curve marked (2) shows the alignment from the second pulse only, which was applied 32.08 ps after the first pulse, and has a peak alignment of 0.67. The solid curve is the measured alignment after both pulses have been applied, and has a peak alignment of 0.74.

Similar in shape and amplitude to this curve. This plot is shown to illustrate the degree of increase in the quality of the revivals. The peak alignment of curve (2) is 0.64, and the peak alignment of the strongest two-pulse curve is 0.68. Even stronger enhancement should be possible by optimizing the aligning pulse intensities and durations. A point of note is the change in the shape of the revivals. When the second pulse is applied as the molecules are approaching antialignment, the two-pulse revival shows strong antialignment and poor alignment, while the revivals formed when the second pulse comes later favour alignment over antialignment.

To illustrate the general applicability of two-pulse alignment, figure 4 shows two-pulse alignment data for oxygen rather than nitrogen. In this experiment, the second pulse is applied not at the first full revival, but at the relatively late revival 32.08 ps, or 2.75 fundamental rotational periods, after the first pulse. The following revival at 35 ps is observed. Alignment in this case made a more dramatic increase from 0.67 to 0.74. The molecular beam in the oxygen experiment was formed by a 100 µm aperture backed by oxygen at 26 Torr and about 40 µm wide. The different molecular beam conditions would result in different initial rotational temperatures, and we expect the wider nitrogen beam would lead to reduced alignment from exposure to more variation in the laser intensity.

We have demonstrated experimental results in which field-free alignment of molecules induced by short laser pulses has been enhanced by using a sequence of two laser pulses. The first laser pulse prepares a rotational wavepacket. When the molecules are in the process of aligning, a second pulse can accelerate the alignment process, leading to higher peak alignment than would occur during a single pulse revival [14].

Stronger alignment has immediate practical implications for improved experiments using aligned molecules, but enhanced alignment is only part of a more general result that rotational revival structures can be modified by using a second laser pulse. Enhanced alignment is one possible effect, but there are infinitely many possible pulse combinations available.

One pulse combination that we have begun studying is the application of the second pulse at a fractional revival, where components of the rotational wavepacket can be addressed.
independently. This freedom may let us coherently switch revivals on and off [16]. Modified revivals have implications not only for alignment, but also for using revivals as a new type of quantum logic system [15].

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References