

Observation of Enhanced Field-Free Molecular Alignment by Two Laser Pulses

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We show experimentally that field-free alignment of iodobenzene molecules, induced by a single, intense, linearly polarized 1.4-ps-long laser pulse, can be strongly enhanced by dividing the pulse into two optimally synchronized pulses of the same duration. For a given total energy of the two-pulse sequence the degree of alignment is maximized with an intensity ratio of 1:3 and by sending the second pulse near the time where the alignment created by the first pulse peaks.

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Experiments on molecules aligned to given space fixed axes are becoming possible due to rapid development of laser based alignment and orientation techniques [1]. In many applications it is desirable and even necessary that the aligned molecules are free of the field that creates their spatial orientation. One way to achieve field-free alignment is to irradiate molecules by an intense nonresonant laser pulse that is short with respect to their rotational periods. If the laser field is linearly polarized the molecular axis with the largest polarizability experiences an impulse towards alignment along the polarization direction. Calculations and recent experiments have shown that field-free alignment can be reached both shortly after the peak of the pulse and at later narrow time windows separated by a molecular rotational period [2–12]. The time dependent alignment results from dephasing and rephasing of the eigenstates in the rotational wave packet formed by the short laser pulse.

For most applications it is also important to ensure that the degree of alignment obtained under field-free conditions is as high as possible. This requires minimizing the rotational temperature of the molecules and selecting the optimal duration and intensity of the alignment pulse. For a given pulse duration the degree of alignment can be improved by increasing the intensity. However, the improvement is often limited by saturation of the alignment process [6,7,13] or by the maximum intensity that can be applied to the molecule without ionizing it. As a way to strongly enhance the degree of alignment of linear molecules Averbukh and Arvieu [14] and Leibscher *et al.* [13,15] recently suggested to employ a train of short laser pulses. The authors found optimal two- and three-pulse alignment schemes, defining the optimal intensity ratios and optimal pulse timings, and showed that the resulting degree of alignment can be strongly improved compared to the alignment induced by a single short pulse [16].

In this Letter we provide the first experimental evidence of enhanced alignment using two optical pulses instead of one. Our studies are carried out on the asymmetric top molecule iodobenzene rather than on a linear molecule, but our findings are in very good qualitative

agreement with the predictions of Leibscher *et al.* We measure the degree of alignment of a sample of rotationally cold iodobenzene molecules at different times following irradiation by either one or two 1.4 ps long, linearly polarized, nonresonant laser pulses. It is shown that the degree of field-free alignment induced by one intense pulse can be significantly improved by replacing it with two pulses containing the same total energy. We find that double pulse alignment is optimized when the second pulse is sent near the time where maximum alignment induced by the first pulse occurs. In addition, for a given total energy of the two pulses the strongest double pulse alignment is achieved if the second pulse is approximately 3 times as strong as the first pulse.

Most of the experimental setup has been described previously [11] and only the important features are discussed here. Briefly, a pulsed molecular beam is formed by expanding 1 mbar of iodobenzene in 3 bars of helium and crossed by three focused laser beams. All laser beams are linearly polarized parallel to each other. The alignment pulses are obtained by passing part of the output from an amplified Ti:sapphire femtosecond laser system ($\lambda = 800$ nm) through a grating stretcher. This allows us to change the pulse duration in the range 100 fs–4.5 ps and also to change the sign of the chirp. In this work we fix the pulse duration at 1.4 ps (FWHM, Gaussian shape, and negative chirp). The output of the stretcher is split into two temporally separated pulses using a Mach-Zender interferometer, where the first beam splitter is replaced by a half wave plate and a polarizing beam splitter. Hereby, the delay between the two pulses can be tuned between 0 and 1000 ps and the ratio between their energies can be varied freely while keeping the total energy constant.

To measure the degree of alignment as a function of time we dissociate the iodobenzene molecules by a 400 fs long pulse ($\lambda = 266$ nm), delayed by a variable time with respect to the alignment pulses, and record the angular distribution of the I photofragments. The photofragments are selectively ionized by resonant (2 + 1)-photon absorption from a 5 ns pulse ($\lambda = 304.59$ nm), sent 12 ns

after the dissociation pulse, and accelerated towards a two-dimensional detector using a weak electrostatic field. Thus, time-resolved images are obtained by recording I^+ ions at different delays between the dissociation pulse and the alignment pulses. By performing Abel inversion of the two-dimensional ion images and correcting for the angular selectivity of the dissociation process we obtain the normalized angular distribution, $f(\theta)$, of the photo-fragments at the time of the dissociation pulse [11,18]. This enables us to determine the conventional measure for the degree of strong laser induced alignment, $\langle \cos^2\theta \rangle = \int f(\theta)\cos^2\theta \sin\theta d\theta$, where θ is the polar angle between the polarization axis of the alignment pulses and the C-I axis of iodobenzene [19].

Figure 1 shows the experimental $\langle \cos^2\theta \rangle$ values at different times obtained with either one or two alignment pulses. When only one pulse is used, centered at $t = 0$ ps, with a peak intensity of 1.4×10^{12} W/cm², the degree of alignment (squares) reaches a maximum of ~ 0.52 around $t = 5$ ps whereupon it gradually decreases to ~ 0.37 at $t = 13.5$ ps. If the intensity is increased by a factor of 3 (4.1×10^{12} W/cm²), corresponding to the second pulse centered at $t = 4.5$ ps, the alignment dynamics becomes faster in agreement with previous short pulse alignment experiments on iodobenzene [11]. The maximum degree of alignment has slightly increased to ~ 0.56 , and although it is reached already at ~ 2 ps after the center of the pulse it is still essentially field-free since the intensity is reduced to $<1/100$ of its peak value [20]. When both the weak first pulse and the strong second pulse are used (filled circles) with the relative timing depicted in Fig. 1, the degree of alignment also reaches a maximum ~ 2 ps after the second pulse ($t = 6.5$ ps), i.e., still under field-free conditions. However, $\langle \cos^2\theta \rangle$ is now ~ 0.66 , which is significantly larger than the 0.56 obtained with the second pulse only. At larger times

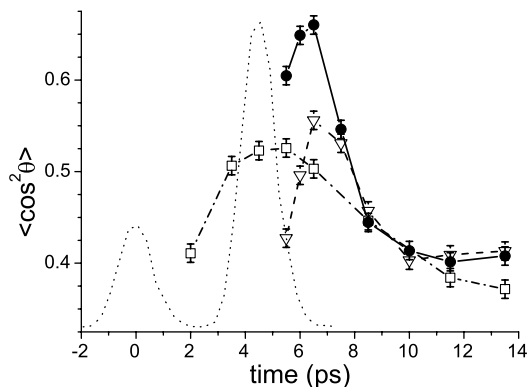


FIG. 1. The degree of alignment vs time for the first pulse only (squares), the second pulse only (triangles), and both pulses (filled circles). The lines connecting the points serve to guide the eye. For a sample of randomly oriented molecules $\langle \cos^2\theta \rangle = 1/3$. The measured intensity shapes of the alignment pulses are given by the dotted line (arb. units).

the degree of alignment drops rapidly due to rotational dephasing.

To test whether the enhanced alignment is simply due to the increased energy of the two pulses Fig. 2 shows a direct comparison of the time dependent alignment obtained with the two-pulse sequence from Fig. 1 and that obtained if the total energy is contained in a single pulse, i.e., an intensity of 5.5×10^{12} W/cm², centered at the same time as the second pulse. It is immediately clear that the two-pulse scheme provides a much stronger maximum alignment. In fact, the highest $\langle \cos^2\theta \rangle$ value of ~ 0.57 obtained with the 5.5×10^{12} W/cm² pulse is only 0.01 larger than that obtained for an intensity of 4.1×10^{12} W/cm² (the second pulse in Fig. 1), indicating that the degree of alignment is saturating in this intensity range. Furthermore, the intensity of the single pulse containing the total energy is so high that multiphoton ionization is starting to appear, which prevents stronger alignment by a further increase of the intensity. Thus, the results of Fig. 2 demonstrate that it is possible to overcome the limitation on the degree of alignment, imposed by the maximum alignment field a molecule can be exposed to without ionizing, by distributing the total energy in two properly timed pulses. This option is not possible in the long pulse (adiabatic) regime where the degree of alignment follows the instantaneous intensity of the alignment field [21].

The enhanced alignment illustrated in Figs. 1 and 2 was obtained by synchronizing the second pulse to near the time where the alignment induced by the first pulse peaks. This conforms with the classical explanation given by Leibscher *et al.* [13,15]: The first pulse forces a large number of the molecules into the angular range where $\sin\theta \approx \theta$. Upon exposure to the second pulse these molecules receive an impulse proportional to θ and, thus, they all align, i.e., reach $\theta = 0$, at the same time. To further investigate the effect of the relative timing between the

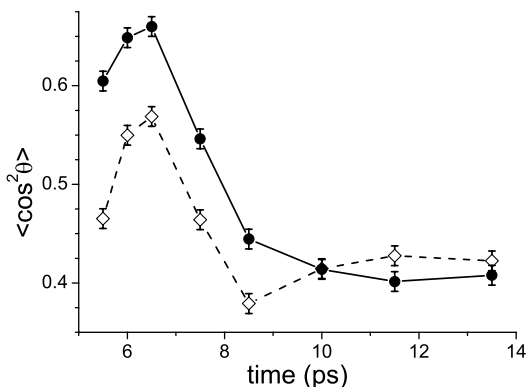


FIG. 2. The alignment dynamics due to a single pulse with intensity 5.5×10^{12} W/cm² (diamonds) and due to the two-pulse sequence illustrated in Fig. 1 (filled circles). The lines connecting the points serve to guide the eye. The total energy of the two-pulse sequence is the same as the energy for the single intense pulse.

two pulses on the best degree of field-free alignment, we measured $\langle \cos^2\theta \rangle$ immediately after the turnoff [22] of the second pulse as a function of the temporal pulse separation. The results obtained for $I_1/I_2 = 1:3$ are displayed in Fig. 3(a) along with the degree of alignment induced by the first pulse at the time when the second pulse is sent [Fig. 3(b)]. The upper curve [Fig. 3(a)] shows that the strongest degree of double pulse alignment is reached when the second pulse is sent approximately 5.5 ps after the center of the first pulse. As seen by comparison with the lower curve [Fig. 3(b)] this is approximately at the time where the alignment due to the first pulse peaks. The qualitative agreement with the optimal timing predicted by Leibscher *et al.* is gratifying considering the fact that their calculations are obtained for linear molecules at zero rotational temperature. Figure 3(a) also shows that when the pulse separation is increased beyond 6 ps the double pulse alignment weakens concurrently with the decrease of the alignment from the first pulse [Fig. 3(b)]. The double pulse alignment dynamics was not investigated for pulse separations lower than 3.5 ps to avoid any effects from the onset of interference as the two alignment pulses starts to overlap temporally.

We also measured the dependence of the field-free double pulse alignment on the relative timing for intensity ratios, other than the 1:3 results shown in Fig. 3, while keeping the total energy of the two pulses constant.

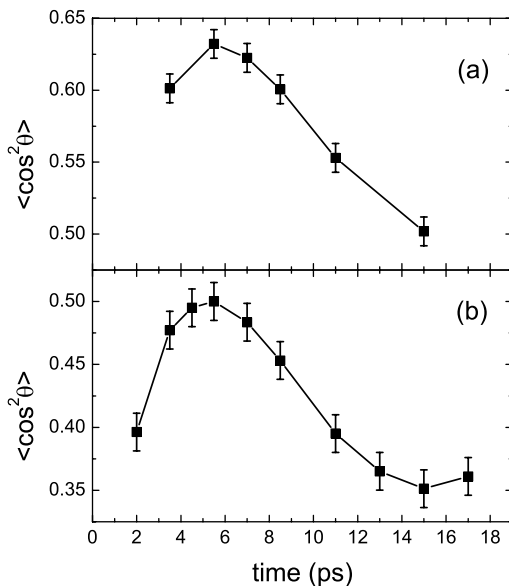


FIG. 3. (a) $\langle \cos^2\theta \rangle$ measured 2 ps after the peak of the second pulse as a function of the time separation between the two pulses. The intensity ratio is 1:3 and $I_1 = 1.4 \times 10^{12}$ W/cm², $I_2 = 4.1 \times 10^{12}$ W/cm². (b) The degree of alignment created by the first pulse only, measured at the time when the second pulse is sent. The data displayed in (a) and (b) were recorded for a higher rotational temperature than the data displayed in Figs. 1 and 2. This explains why the overall degree of alignment is lower in Fig. 3 than in Figs. 1 and 2.

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In all cases we found that the optimal field-free alignment occurs approximately 2 ps after the peak of the second pulse and when the second pulse is sent at or slightly after the alignment maximum due to the first pulse. These observations are summarized in Fig. 4 where the optimal degree of alignment at different I_1/I_2 ratios is displayed. The figure shows that when the intensity of one pulse containing the total energy ($I_1 = 0$, $I_2 = 5.5 \times 10^{12}$ W/cm²) is approximately at the onset of ionization, a better field-free alignment can be obtained by dividing the total energy into two, optimally synchronized pulses with $I_1/I_2 \leq 1$. In particular, the strongest alignment is obtained when the second pulse is approximately 3 times more intense than the first pulse. A direct comparison with the results of Leibscher *et al.* is not possible. However, they show that for molecules with zero rotational temperature optimal double pulse alignment can be achieved with an intensity ratio of approximately 1:10, but as the temperature is increased, enhanced alignment requires a more intense first pulse with a value that approaches that of the second pulse [15].

Our previous studies of nonadiabatic alignment of iodobenzene and other asymmetric top molecules with one pulse have shown that the first maximum in the degree of alignment shortly after the pulse turnoff is stronger than the subsequent maxima reached at the rotational revivals [11,23]. This motivates why we choose the small pulse separation of ~ 5 –8 ps in our double pulse experiment rather than the several hundred ps that would correspond to a full rotational period [11,23]. By contrast, in the work of Leibscher *et al.* on linear molecules the optimal separation is close to half a rotational period since the global alignment maximum induced by the first pulse occurs at the first half revival [13,15].

In the present work we restricted our studies of the double pulse alignment dynamics to shortly after the second pulse. Preliminary measurements around the first

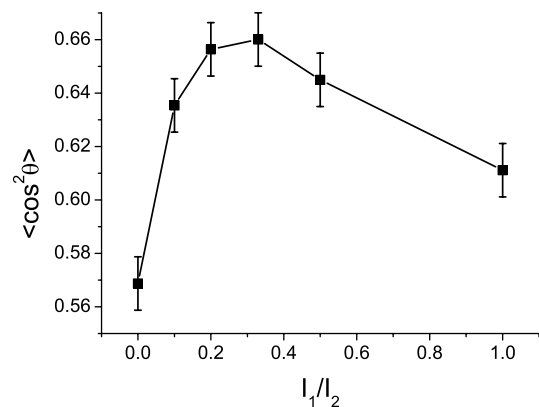


FIG. 4. The optimal degree of field free double pulse alignment, measured 2 ps after the peak of the second pulse, as a function of the intensity ratio, I_1/I_2 , for a fixed total energy. For $I_1/I_2 = 0$, $I_2 = 5.5 \times 10^{12}$ W/cm², and for $I_1/I_2 = 1$, $I_1 = I_2 = 2.6 \times 10^{12}$ W/cm².

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(C-type) revival at ~ 378 ps did not reveal stronger alignment than the maximum observed 2 ps after the second pulse. The possibility of defining optimal two (or multi)-pulse conditions for enhanced alignment on the revivals is, however, inviting and as yet unexplored.

The present experimental demonstration of alignment enhancement by two pulses instead of one employs 1.4-ps-long pulses. Experimentally, we did not test if other pulse durations could further improve the degree of alignment of iodobenzene under the action of two pulses, but we note the following. If shorter pulses are used it is necessary to lower the total energy to avoid multiphoton ionization. Since the multiphoton ionization rate has a highly nonlinear dependence on the intensity it will be necessary to lower the energy more than just proportional to the lowering of the duration. As a result the total impulse exerted on the molecule is lowered and, therefore, improved alignment seems unlikely. In the short pulse limit a more promising alternative seems to split up the total energy in more than two pulses [15]. Conversely, if the pulse duration is increased the total energy can be increased. This might create stronger alignment, but the larger pulse duration could imply that the first alignment maximum is reached before the field is terminated. It is possible that fast pulse truncation by pulse shaping can overcome this problem.

In summary, using time-resolved photofragment imaging we demonstrated that the strongest field-free alignment of iodobenzene molecules obtained by a single 1.4-ps-long pulse can be significantly improved by dividing the pulse into two properly timed subpulses. The strongest double pulse alignment occurs shortly after the turnoff of the last pulse, and it is optimized when the second pulse is approximately 3 times as intense as the first pulse and synchronized to the time where the maximum alignment due to the first pulse occurs.

The potential for further enhancement and control of molecular alignment and orientation through application of pulse trains, and more generally, optimally shaped laser pulses, seems very large and promising [17,24,25].

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Note added.—Related work on two-pulse alignment has been reported very recently [26]

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