Effects of aligning pulse duration on the degree and the slope of nitrogen field-free alignment*

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Through theoretical analysis, we show how aligning pulse durations affect the degree and the time-rate slope of nitrogen field-free alignment at a fixed pulse intensity. It is found that both the degree and the slope first increase, then saturate, and finally decrease with the increasing pump duration. The optimal durations for the maximum degree and the maximum slope of the alignment are found to be different. Additionally, they are found to mainly depend on the molecular rotational period, and are affected by the temperature and the aligning pump intensities. The mechanism of molecular alignment is also discussed.

Keywords: field-free molecular alignment, femtosecond phenomenon, strong filed physics

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1. Introduction

Molecular alignment induced by a strong non-resonant laser field was initiated by the work of Friedrich and Herschbach[1] and Seideman[2] in the 1990s. Since then, such alignment has been widely studied both theoretically[3] and experimentally.[4−6] There are two kinds of molecular alignment. When the aligning pulse duration is longer than the molecular rotational period, the molecular alignment occurs at the peak of the aligning laser field, and this is called adiabatic alignment.[4] The coexistence of the aligning pump limits the generality of this method. In contrast, if the aligning pump duration is far shorter than the molecular rotational period, the molecules receive a rapid momentum kick from the pulse and start to align towards the polarization direction of the laser field. Since this alignment repeats with a certain rotational period even after the laser effect, it is called field-free alignment[5,6] and is relatively easy to apply.

The alignment degree is characterized by the average $\langle\cos^2\theta(t)\rangle$, where $\theta(t)$ is the angle between the laser polarization direction and the molecular axis and can be measured by using many experimental techniques.[7,8] Pre-aligned molecules are important for strong field ionization and high-order harmonic generation,[9−13] where the degree of alignment is directly related to the structural resolution. An increased angular resolution facilitates the comparison of experimental results with theory. Additionally, the molecular index will be modulated once the molecules have been aligned. This has already been reported and applied for large tunable few-cycle femtosecond laser pulse generation. The time-rate slope of the field-free alignment has been demonstrated to be the key factor for obtaining a wider frequency tuning range of a probe pulse.[14,15]

Since the first experimental achievement of molecular alignment,[4] researchers have searched for factors that affect the alignment degree and slope. Those commonly used parameters include the pulse intensity[16] and the temperature.[17,18] The use of multiple aligning pumps has recently been demonstrated.[19−23] And it has been reported that a special phase-and-amplitude-shaped femtosecond laser pump can be used to achieve the optimized alignment.[24,25] Among those factors, the role of aligning pulse durations is rarely discussed, the only finding known to us is that the alignment degree increases with the increasing pulse energy when the duration is far shorter than the rotational period. As the pulse intensity is limited by the ionization, it is better for us to know how the pump duration affects the degree and the slope of the field-free alignment at a fixed intensity. Is there an optimal duration for achieving the maximum degree and slope of alignment? What factors govern this best pump duration? All these issues are discussed for N_2 in this work. In Section 2,

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we give simulation results at room temperature. In Section 3, we discuss results obtained at lower temperature.

2. Optimal pulse durations at room temperature

As previously stated, $\langle\cos^2\theta(t)\rangle$ is the thermally averaged quantum value usually used to characterize the degree of alignment. The value of $\langle\cos^2\theta(t)\rangle$ and its time-rate slope can be calculated by solving the following Schrödinger equation:[26]

$$\mathrm{i}\frac{\partial}{\partial t}\psi(\theta,t) = \Big[\frac{J^2}{2I} - \Delta\alpha\pi|E_{\mathrm{pump}}(t)|^2\cos^2\theta(t)\Big]\psi(\theta,t), \quad (1)$$

where J is the angular momentum operator, I is the moment of inertia, and $|E_{\mathrm{pump}}(t)|$ is the envelope of the x-axis-polarized pump pulse that aligns the molecules.

Besides the pulse duration, the other important characteristic parameter of the intense laser pulse is the pulse energy or the peak intensity. And these two parameters are dependent on each other. To thoroughly investigate how the duration of the aligning pump pulses affects the molecular alignment degree and its slope, the other parameter should be fixed first. The relationships between the pump duration and the induced maximum degree of alignment are obtained for different fixed pulse energies and different fixed pulse intensities, as shown in Figs. 1(a) and 1(b), respectively.

It is normally believed that the maximum alignment degree is determined by the pulse energy when the pulse duration is far less than the molecular rotational period. However, as shown in Fig. 1(a), once the pulse duration exceeds 1/200 of the rotational period, which is about 42 fs here for N_2, the alignment degree is no longer the same as that for the shorter durations and decreases even though the same pump energy is applied. This implies that the duration of the aligning pump pulses starts to affect the alignment degree when it exceeds 1/200 of the rotational period. The results in Fig. 1(b) tell us that at a certain intensity, when the laser pulse broadens, the degree of N_2 alignment first increases, then saturates, and finally decreases. Additionally, it is clear from the curves in Fig. 1(b) for different intensities that the degree of alignment increases with the increasing pulse peak intensity. This is consistent with the results in our previous report.[27] The alignment curves for different intensities tell us that the optimal pulse durations for the highest alignment degree become slightly shorter as the intensity of the aligning pulse increases. But they are all around the value of 250 fs. This optimal pulse duration at room temperature is about 3.0% of the rotational period of N_2, which is 8.4 ps.

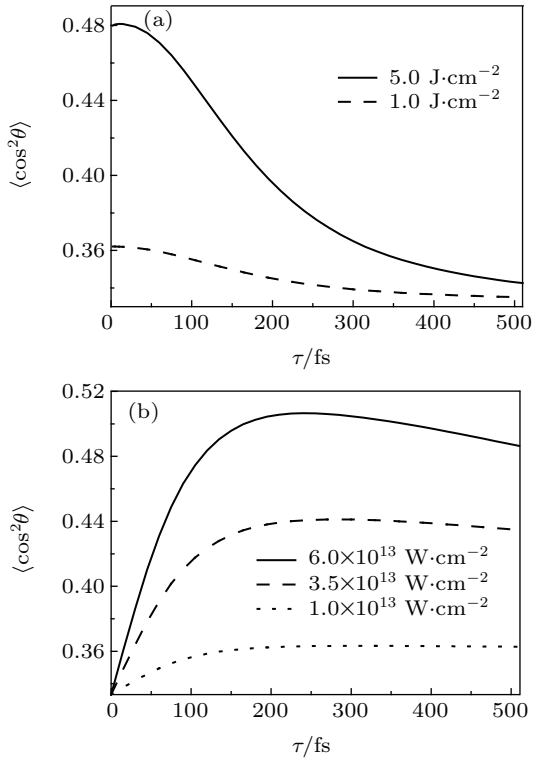


Fig. 1. Relationships between the maximum degree of alignment and pulse duration τ at room temperature for (a) different fixed pulse energies and (b) different fixed pulse intensities.

In the application of tuning the frequency of the probe pulse,[13,14] it is the time-rate slope of the alignment that determines the tuning range. The alignment slopes at the full rotational period versus the pump duration are shown for different pulse energies and different pulse intensities in Figs. 2(a) and 2(b), respectively. Similar to the results in Fig. 1(a), once the pulse duration exceeds 1/200 of the rotational period, which is about 42 fs for N_2, the slope in Fig. 2(a) is no longer the same as that for the shorter durations. The relationship between the pump duration and the induced alignment slope shown in Fig. 2(b) has the same pattern as that for the induced alignment degree. And the steepest slope is achieved at a particular pulse duration, which is weakly decreased with the increase of the pulse intensity and is about 120 fs at room temperature (about 1.5% of the rotational period of N_2). However, when comparing the results in Figs. 1(b) and 2(b), the optimal pulse durations

for the maximum degree and the maximum slope of alignment are clearly different, even though the same pulse intensity and the same molecular condition are considered. This means that the best aligning pump for the highest alignment degree does not guarantee the widest tuning range of the probe pulse.

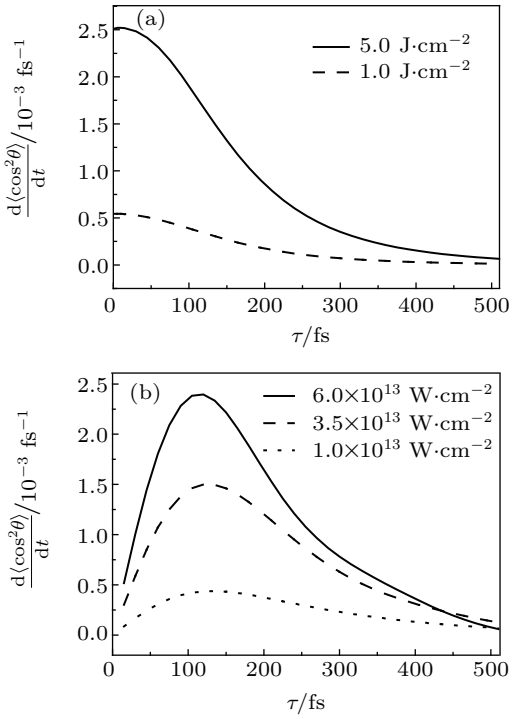


Fig. 2. Alignment slopes at the full rotational period versus pulse duration τ at room temperature for (a) different pulse energies and (b) different pulse intensities.

To understand the limitations of the aligning pulse durations, we study how the pump-induced N_2 alignment varies with time, and the results for a fixed intensity of 3.5×10^{13} W·cm^{-2} are shown in Fig. 3. Different curves in the figure are for different pulse durations. Two characteristics can be found in the figure. First, considering all alignment curves, we can see that all curves first increase, then saturate, and eventually decrease as the pump duration increases. The strongest alignment appears at the pulse duration of about 255 fs for the fixed intensity of 3.5×10^{13} W·cm^{-2}. This is consistent with the results shown in Fig. 1(b). Second, we focus on a single alignment curve for a certain pulse duration, the first peak of transient alignment is obtained at a certain time point, such as 85 fs on the solid curve (the intersection point of the solid curve and the vertical line). The transient alignment increases with time before this point, and decreases after it. For the moment, we call this point the optimal time point for the first peak of transient alignment and denote it t_0. The optimal time point is determined by the distribution of the rotational wave-packet. The higher rotational eigenstates the distribution covers, the shorter the time needed to achieve the first peak. The peaks of the alignment curves in Fig. 3 shift forward when the pump duration increases, because the higher energy excites higher rotational eigenstates. However, this shift is extremely small for pulse durations shorter than 255 fs, and their optimal time coincides around 85 fs as shown in Fig. 3. But once the pump duration exceeds 255 fs, the shift becomes obvious (e.g., the alignment curves for 315 fs and 405 fs in Fig. 3).

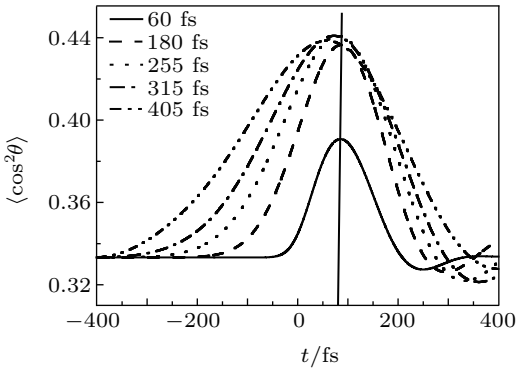


Fig. 3. Alignment degrees versus time for different pump durations.

As discussed above, the optimal time point for the first peak of the transient alignment is determined by the distribution of the rotational wave-packet, and it is shorter when the higher rotational eigenstates are excited. Complementary to the above results, the pulse duration is fixed at 180 fs, and we show the relationship between the transient alignment and the pump intensity in Fig. 4. The intersection points between the three curves and the vertical line tell us that the optimal time point for the first peak of the transient alignment is slightly affected by the pump intensity.

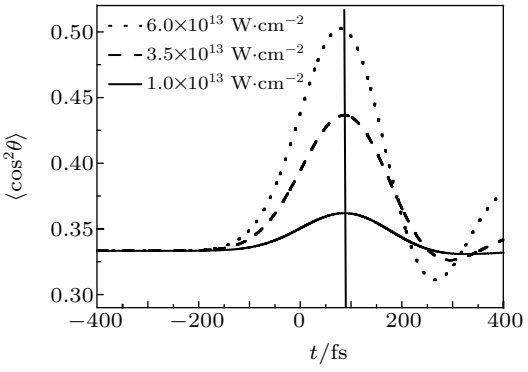


Fig. 4. Alignment degrees versus time for different pump pulse intensities, all pulse durations are 180 fs.

To obtain a better understanding of the molecular alignment, the transient molecular alignments are

shown in Fig. 5, together with their corresponding pump intensities. It is known that the integration area of the intensity curve represents the laser pulse energy. Additionally, the revival variation of the field-free alignment tells us that once the transient alignment reaches its first peak, the following is only a periodic repetition. This means that only the pulse energy before the optimal time t_0 contributes to the maximum degree of alignment. The pulse energy after this time does not affect the maximum degree of alignment, however it delays the minimum alignment as shown in Fig. 5 and decreases the alignment slope at the full rotational period (the dashed line in Fig. 2(b)). This is the reason why the optimal pump duration for the maximum alignment slope (about 120 fs in Fig. 2(b)) is shorter than that for the maximum alignment degree (about 250 fs in Fig. 1(b)) for the fixed intensity of 3.5×10^{13} W·cm^{-2}.

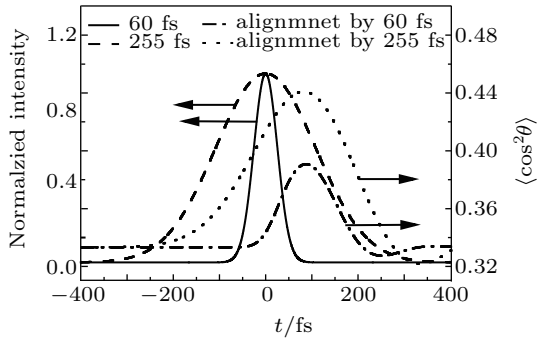


Fig. 5. Alignment degrees versus time for different pump pulse durations, together with their corresponding pump intensities, all peak intensities are 3.5×10^{13} W·cm^{-2}.

3. Results for lower temperature

It is concluded from Section 2 that irrespective of the peak intensity, there are two special pump pulse durations, one for the maximum degree, and the other for the maximum slope of N_2 alignment. Meanwhile, the transient alignment results show that the first peak of transient alignment can be obtained at an optimal time point t_0, which is mainly determined by the molecular rotational period. However, the transient alignment can still be affected by temperature, because it is the temperature that decides the initial rotational eigenstates. So, how the temperature affects the alignment and time t_0 is studied in this section. For all the results below, the temperature is set at 90 K.

Similar to the results in Fig. 1(b), the relationships between the pump duration and the induced maximum degree of alignment at different pulse intensities are shown in Fig. 6. The variational pattern is still the same. However, here in a cooler environment, the alignment degree is maximized at a pulse duration of about 300 fs for the fixed intensity of 3.5×10^{13} W·cm^{-2}, which is longer than that (255 fs) at room temperature. Meanwhile, figure 7 gives the relationship between the alignment slope at the full rotational period and the pump duration. The results are the same as those obtained at room temperature in Fig. 2(b), and the pulse duration for the largest slope is different from that for the largest degree. It is also shown in Figs. 6 and 7 that as the peak intensity increases, the optimal pulse durations for the maximum degree and the maximum slope of alignment become much shorter at lower temperature.

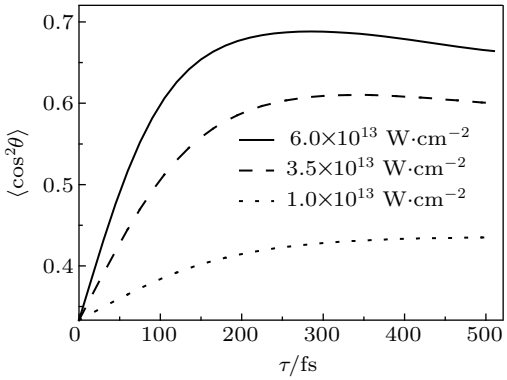


Fig. 6. Relationships between the maximum degree of alignment and pulse duration τ at 90 K for different pulse intensities.

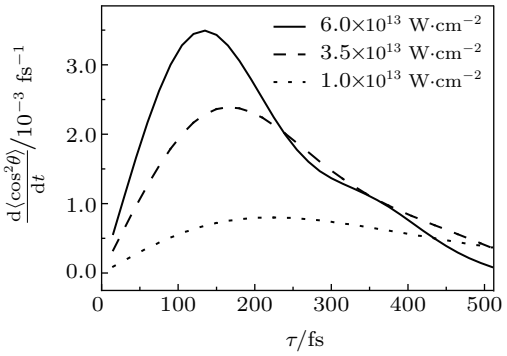


Fig. 7. Alignment slopes at the full rotational period versus pulse duration τ at 90 K for different pulse intensities.

Figure 8 presents the results of transient alignment induced by aligning pumps of different durations, where their peak intensities are all fixed at 3.5×10^{13} W·cm^{-2}. The vertical line tells us that the optimal time point t_0 for the first peak of the transient alignment is about 120 fs at 90 K, which is longer than that at room temperature. This is because the rotational wave-packet at lower temperature covers fewer rotational eigenstates. Therefore, the molecules rotate more slowly and achieve the first peak much later.

This is also the reason why the best pulse duration for the maximum degree of alignment at 90 K (about 300 fs as shown in Fig. 6) is longer than that at room temperature (about 250 fs as shown in Fig. 1(b)) for the fixed intensity of 3.5×10^{13} W·cm^{-2}.

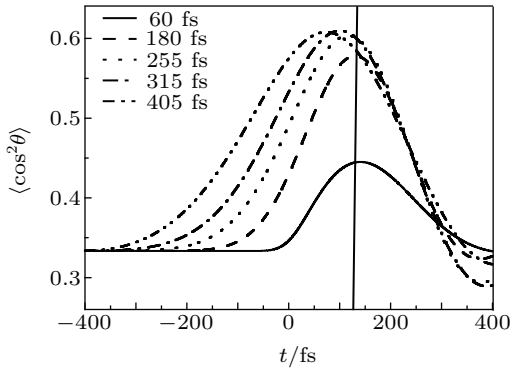


Fig. 8. Alignment degrees versus time for different pulse durations at 90 K, all the peak intensities are 3.5×10^{13} W·cm^{-2}.

When comparing the degree and the slope of alignment at room temperature and 90 K, the enhancement of the alignment slope in the cooler environment (Fig. 9(b)) is not as strong as that of

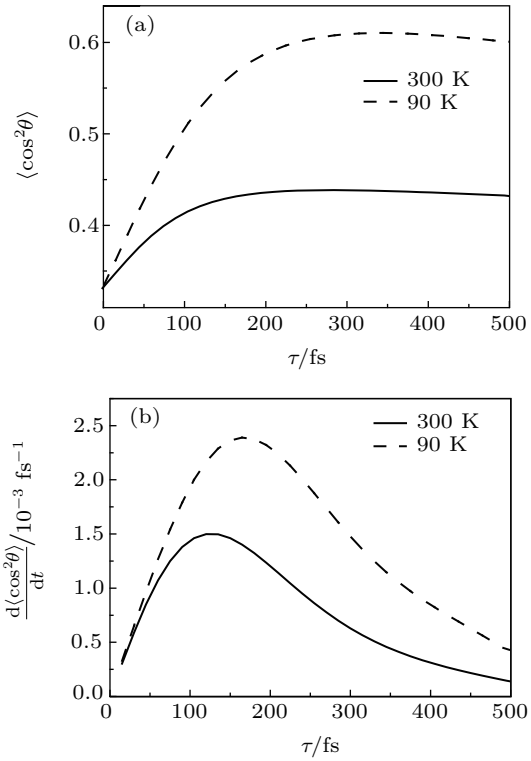


Fig. 9. (a) The maximum alignment degree and (b) the alignment slope at the full rotational period versus pulse duration τ. All the peak intensities are 3.5×10^{13} W·cm^{-2}.

the alignment degree (Fig. 9(a)). This is because the enhancement of the alignment slope is determined not only by the alignment degree but also by the time range that the nearly linear alignment covers. The transient alignment results at around the rotational period in Fig. 10 show that the time range decreases with temperature, which is consistent with the result that the alignment degree needs more time (greater t_0) to reach its first peak at the lower temperature. Therefore, we can take advantage of the lower temperature to remarkably enhance the alignment degree. A lower temperature is also helpful for the alignment slope, but the enhancement ratio is smaller.

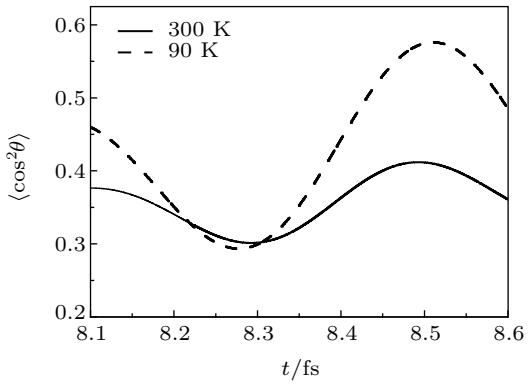


Fig. 10. Transient alignment results at around the full rotational period (8.4 ps for N_2), the pump intensities are 3.5×10^{13} W·cm^{-2}, and the durations are 255 fs.

4. Conclusion

We theoretically analyze how the aligning pulse durations affect the degree and the time-rate slope of N_2 field-free alignment. It is found that at a fixed intensity, both the degree and the slope of the N_2 alignment first increase, then saturate, and finally decrease with the increasing pump duration. At room temperature, the best pump duration for the maximum degree of alignment is about 3.0% of the rotational period, and it is about 1.5% of the rotational period for the maximum slope. These best durations are both limited by time t_0, at which the first maximum alignment is obtained. They are found to decrease with the increases of the temperature and the pump intensity. Also, it is shown that as the temperature becomes lower, both the degree and the slope of alignment increase.

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