Pulse polarization evolution and control in the wake of molecular alignment inside a filament

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Abstract: The polarization evolution and control of a femtosecond laser pulse in the wake of molecular alignment inside a laser filament was investigated. A weak probe pulse was delayed with respect to the field-free revivals of the pre-excited rotational wave-packets created by an infrared filamenting pulse in nitrogen gas. 30° was set between the pump and probe’s initial linear polarization directions in order to control the output probe’s polarization ellipse. The detailed physical response of the probe’s polarization states was analyzed in the wake of alignment and dephasing of molecular N2. The probe’s polarization was modulated by varying the retarded time between the pump and probe pulses.

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References and links

1. Introduction
Filamentation induced birefringence in isotropic gas media has attracted increasing attentions owing to its remarkable effects on the spatial and spectral reshaping of a probing pulse [1–4]. In atomic gases, the ultrafast birefringence was quantified by measuring the filament-induced polarization change of a probe pulse [5–8]. In molecular gases, besides this instantaneous electronic response, a delayed birefringence was induced by the molecular alignment and revivals. The molecular alignment induced birefringence could be measured by means of the weak-field probe polarization technique with 45° between the initially linearly polarized pump and probe pulses [9, 10] or by making use of the spatial cross-defocusing effect of the probe pulse through fixing the pump and probe polarization crossed with each other [11, 12].

P. Béjot et. al [5] has elaborated the pressure (de-phasing) dependence of the polarization diagram of a probe beam driven by an ultrashort laser filament in an argon cell, with an initial angle of 45° between the linear polarizations of the pump and the probe. More recently, the ultrafast birefringence was quantified in argon by measuring the filament-induced polarization rotation of a probe pulse [6]. However, detailed measurement of the response of the probe’s polarization states caused by molecular alignment induced delayed birefringence has been left aside to date. In this paper, the filament was generated by focusing an infrared pump pulse in nitrogen gas. In the wake of the filament, the field-free revivals of the rotational wave-packets of molecular N₂ were analyzed by a weak probe pulse. The detailed physical responses of the output probe’s polarization states were measured at different revivals of molecular N₂. Consequently, the polarization property of the probe pulse could be controlled.

2. Experiment setup
The experimental setup is shown in Fig. 1. A single ~1-cm-long filament was created by focusing a 0.6 mJ, linearly polarized, 60 fs, 800 nm laser pulse in 1 bar of nitrogen gas with a lens (f = 50 cm). The filament was probed by a second linearly polarized, weak pulse (~60 fs, 0.8 μJ, 400 nm). The pump and probe beams were combined together by a dichroic mirror. A delay stage was placed on the pump beam path in order to change the relative time delay between the pump and probe pulses. After the filament, the probe pulse was analyzed by a polarizer and the resulting transmission was projected on a white paper screen. A CCD camera was used to detect the probe scattering from the paper [6, 7]. Here the signal was measured by a CCD camera rather than a photodiode, since we were also trying to investigate the alignment-induced spatial (de)focusing effects during our experiment. The experiment was done by imaging the probe scattering while rotating the transmission axis of the analyzer at different relative delay between the pump and the probe pulses.
During the experiment, the initial polarization direction of the pump pulse was set at 30° to that of the probe in order to induce the evolution of the output probe pulse’s polarization state (this part will be discussed in the following part). This scheme is different from the schemes of the weak field polarization technique and cross-defocusing technique. In the case of the weak field polarization technique, the polarization of the probe is set at 45° to that of the pump. Thus the probe’s polarization has the same amount of projection parallel and perpendicular to the pump polarization direction (optical axis). Therefore, the main axis of the probe will not rotate, if we do not consider spatial effect inside a filament [13]. In the case of cross-defocusing technique, the polarization of the probe pulse is orthogonal to that of the pump (direction of optical axis). Hence its polarization will basically not change. The current scheme provides an alternative approach for the time dependent polarization control.

3. Theory

In this paper, we assume that the propagation wave vectors are in the z direction and the pump linear polarization is along the x direction (0°). For molecular alignment induced birefringence of the probe pulse, the refractive indices’ modulation parallel ($\delta n^{align}_x$) and perpendicular ($\delta n^{align}_y$) to the pump laser field are defined as

$$\delta n^{align}_x(r,t) = 2\pi(N / n_0)\Delta\alpha<\cos^2\theta>_{\tau} - 1/3$$

and

$$\delta n^{align}_y(r,t) = -\pi(N / n_0)\Delta\alpha<\cos^2\theta>_{\tau} - 1/3$$

respectively [14, 15], where N and $n_0$ denote the molecular number density and linear refractive index of the initially randomly aligned molecules. $\Delta\alpha$ is the molecular polarizability anisotropy, $\theta$ is the angle between the pump laser’s polarization and the molecular axis, $<\cdot>_{\tau}$ is the time-dependent ensemble average [7, 16, 17], and $t$ is the retarded time between the pump and probe pulses. The laser-induced plasma defocusing has no preference parallel and perpendicular to the laser field, thus the resulting birefringence $\Delta n^{align}(r,t)$ can be expressed as

$$\Delta n^{align}(r,t) = \delta n^{align}_x(r,t) - \delta n^{align}_y(r,t) = 3\pi(N / n_0)\Delta\alpha<\cos^2\theta>_{\tau} - 1/3.$$

The alignment induced birefringence $\Delta n^{align}(r,t)$ is proportional to $<\cos^2\theta>_{\tau} - 1/3$. The simulated statistic metric $<\cos^2\theta>_{\tau} - 1/3$ for N$_2$ molecules is shown in Fig. 2(a), with the parameters expressed according to the experimental conditions: gas temperature $T = 294$.
K, the peak intensity of the aligning field $I = 5 \times 10^{13}$ W/cm$^2$ and its FWHM duration $t = 60$ fs. Figure 2(b) illustrates the revival of the pre-aligned N$_2$ molecules, which is experimentally measured using the weak field polarization technique [9]. By varying the retarded time between the pump and the probe, the x and y axes become fast and slow axis alternately.

Fig. 2. (a) The calculated revival for molecular N$_2$ ($<\cos^2 \theta> - 1/3$) along the field polarization of the probe pulse. (b) Experimentally measured molecular alignment signal of N$_2$. (c) The polarization scheme for the probe. $\phi$ is rotation angle of the probe.

Due to the molecular alignment induced birefringence, integrated over the whole interaction length (roughly the length of the filament), the probe’s nonlinear phases along the 0° and 90° axes are accumulated separately over the interaction length. By approximating the filament as a uniform cylinder of length $L$, the overall de-phasing $\epsilon(t)$ accumulated between the probe’s two components along the 0° and 90° axes is given by [15]

$$\epsilon = \Delta n_{\text{align}} \omega_0 L / c_0 = 3 \pi \omega_0 L \Delta \alpha(N / n_0 c_0) <\cos^2 \theta> - 1/3 = \eta <\cos^2 \theta> - 1/3,$$

where $\omega_0$ is the laser central angular frequency, $c_0$ is the speed of light in vacuum and $\eta \sim 3 \pi \omega_0 L \Delta \alpha(N / n_0 c_0)$ is a constant. The alignment induced de-phasing $\epsilon(t)$ is proportional to $<\cos^2 \theta> - 1/3$.

As the time delay between the pump and probe varies, the probe pulse will see different birefringence $\Delta n_{\text{align}}(t)$, giving rise to the polarization rotation of the probe and making the initially linearly polarized probe elliptical [5, 6, 8]. A laboratory coordinate system shown in Fig. 2(c) is used to describe the two components of the probe field along the x and y axes.
Under the plane wave assumption, the rotation angle $\phi$ ($\phi = \psi_{\text{probe}}^{\text{input}} - \psi_{\text{probe}}^{\text{output}}$) of the probe can be derived from [6]

$$\tan 2(\psi_{\text{probe}}^{\text{input}} - \phi) = 2E_x E_y \cos \varepsilon / (E_x^2 - E_y^2),$$

(3)

where $E_x$ and $E_y$ are the initial electrical field of the probe projected along (0°, x axis) and perpendicular (90°, y axis) to the pump polarization direction, $\psi_{\text{probe}}^{\text{input}} = \arctan(|E_x| / |E_y|)$ is defined as the angle between the initial linear polarization of the probe and that of the pump (0°, x axis). Throughout this paper, we call it initial polarization angle of the probe. $\psi_{\text{probe}}^{\text{output}}$ is defined as the output polarization angle of the probe, which is actually the angle between x axis and the major axis of the output probe’s polarization ellipse. $\phi$ is positive, when the final polarization direction of the probe rotates clockwise from the initial one with respect to the observer looking into the probe’s propagation direction. Equation (3) describes the rotation of the final probe's polarization. In Eq. (3), the de-phasing term $\varepsilon$ is inside a cosine function. Thus, the relative sense of rotation only depends on the initial pump-probe polarization scheme and the absolute value of de-phasing $\varepsilon$ (not the sign of the de-phasing $\varepsilon$). In case of aligned molecules, it means that the polarization rotation angle $\phi$ is related to the degree of the alignment, not the orientation of the molecules (whether probe sees the in-phase or out-of-phase revivals).

Fig. 3. Movies of the output polarization diagrams of a probe beam under each value of dephasing, with 45° (see Media 1) (a) and 30° (see Media 2) (b) between the initial polarization direction of the probe and the pump.

In order to have a detailed physical picture of the evolution of the probe's polarization, the probe's transmission is calculated by assuming that the output probe beam passed through a cubic polarizer with different values of de-phasing ($\varepsilon$). The output polarization diagrams of a probe beam under different value of birefringence are shown as the movies in Figs. 3(a) and 3(b). The movies display, for each value of de-phasing, the simulated signal of the output probe beam as a function of the orientation of a polarizer. In Figs. 3(a) and 3(b), the ellipticity $\varepsilon$ of the probe polarization is defined as the ratio between the minor and major axis in the probe's polarization diagram. Figure 3(a) exhibits the evolution of the probe's polarization states by using the initial pump-probe polarization scheme which is used by the weak filed polarization technique $\psi_{\text{probe}}^{\text{input}} = 45^\circ$. In Fig. 3(a), by varying the dephasing, the polarization ellipse of the initially linearly polarized probe expands gradually,
until $\epsilon = \pi/4$, it becomes circular. Besides, as the de-phasing changes, the probe's polarization ellipse does not rotate. While the major and minor axes exchange, when the de-phasing $\epsilon$ equals to an integral multiple of $\pi$. While in case of $\psi_{\text{probe}} = 30^\circ$ in Fig. 3(b), by continuously increasing the value of the de-phasing, the polarization ellipse rotates clockwise and back. It is important to note that since our calculation does not consider plasma or molecular effect, the diagrams in Figs. 3(a) and 3(b) cannot depict the output probe's polarization ellipse quantitatively. However, it offers us an intuitive picture of the evolution tendency of the probe's polarization states.

4. Results and discussions

The experiment was carried out by rotating the transmission axis of the analyzer at different pump-probe delay while measuring the probe transmission. We scanned the pump-probe delay over the full range of $N_2$'s molecular revival. As an example, we show one set of the polarization changes $\phi$ of the probe around the full revival region [around 8 ps in Fig. 2(a)] when the molecules evolve from being perpendicular to the pump's linear polarization to being parallel. This evolution is compared with the probe's polarization state when the molecules are randomly oriented. The polarization direction of the initially linearly polarized probe was fixed at $30^\circ$ with respect to that of the pump ($0^\circ$) ($\psi_{\text{probe}} = 30^\circ$). The probes' polarization states around the full revival at time delays B-E as

Fig. 4. Polarization analysis of the transmitted probe in the wake of the pump for initially $30^\circ$ between the pump and probe polarization direction at different delay position as labeled in Fig. 2(a). Black-dotted lines correspond to probe polarization state at delay-A in Fig. 2(a), while the Red-dotted lines correspond to probe polarization state at delays B-E in Fig. 2(a). Black dash line is at $30^\circ$ and red dash lines indicate the main axis of the probe polarization. $\phi$ is rotation angle of the probe.
labeled in Fig. 2(a) are shown in Figs. 4(a)-4(d) (red curve) as compared to the probe’s polarization state (black curve) at delay A in Fig. 2(a). Delay A is for random orientation of the molecules which is equivalent to the initial polarization of the probe; delays B-E give the evolution of the molecules from perpendicular to parallel. In Figs. 4(a)-4(d), the rotation angle is the angle between the initial probe polarization direction (30°) and the final direction for the major axis of the probe’s ellipse. The measured ellipticity is the ratio between the length of the minor axis and that of the major axis on the experimentally measured angular transmission. We compare the probe’s polarization states for various states of molecular alignment [red curves in Figs. 4(a)-4(d)] with the one corresponding to randomly distributed molecules [black curves in Figs. 4(a)-4(d)]. We see that after propagating through the alignment region in the wake of the pump, the probe polarization integrated over the whole beam became elliptical, with the major axes rotating clock-wise as compared with the initial polarization direction.

Fig. 5. Rotation angle of the major axis (red circle) (a) and the ellipticity (red circle, solid line) (b) of the probe’s elliptical polarization as a function of the relative delay between pump and probe. Numerical simulation (black curve) at 296 K for a Gaussian pump envelope of 80 fs pulse duration.

The polarization rotation angles $\phi$ and the ellipticity $e$ of the output probe as a function of delay over the revivals of molecular N$_2$ are summarized in Figs. 5(a) and 5(b), respectively. The theoretical calculation is done by using the calculated de-phasing $\varepsilon = 3\pi\omega_L \Delta \alpha (N / n_0 c_0) \langle \cos^2 \theta \rangle$, to retrieve the rotation angle $\phi(t)$ and the ellipticity $e$ of the probe. The parameters are chosen similar to the experimental conditions: polarizability anisotropy of $\Delta \alpha_{N_2} = 0.93 \times 10^{-24}$ cm$^3$, and molecular number density $N_{N_2} = 2.5 \times 10^{19}$ cm$^{-3}$ [14]. A filament length of 1.3 cm is used for better fitting the experimental curve in Fig. 5(a). The result of the theoretical calculation is shown as the black curve in Figs. 5(a) and 5(b). When $\psi_{\text{input}} = 30^\circ$, Eq. (3) gives

$$\phi(e_i) = -0.5 \arctan(\sqrt{3} e_i) + 30^\circ$$  \hspace{1cm} (4)
with the de-phasing $\varepsilon_i$ inside a cosine function. In Eq. (4), the rotation angle of the probe $\phi(\varepsilon_i)$ increases with the absolute value of de-phasing $|\varepsilon_i|$ monotonically. Thus the delay time dependent rotation angle $\phi(t)$ actually follows $<\cos^2 \theta, -1/3>_{\varepsilon_i}$. The experimental results are nicely reproduced by the simulation. In the wake of a filament, the main axis of the probe’s polarization ellipse can be manipulated by adjusting the time delay between the pump and probe pulse. It might have further applications on techniques like polarization gating and remote control.

In Fig. 5(b), with $\psi_{\text{input}} = 30^\circ$, the experimentally measured and theoretically calculated ellipticity has qualitatively similar time delay dependent modulation. The experimental results are larger than the simulation results. One possible reason could be that our simulations are based on plane wave assumption, in which the plasma and molecular effect are not considered. While in the experiment, the aligned molecules trends to diffract perpendicular-to-pump component outside the central core of the probe [13]. This part of the probe would no longer see the aligned region and finally experience different birefringence as compared with the output probe within the central core, which possibly induces the difference of the ellipticity.

5. Conclusions

The evolution of the probe’s elliptical polarization states as a result of molecular rotation through Raman-type excitation as well as the birefringence of the rotational wave-packet were measured experimentally at various revival times. Different initial polarization scheme was used in order to control the output probe's polarization ellipse. The detailed physical responses of the probe’s polarization states were analyzed and predicted. Besides, the experimental results provide a field-free, broadband control in molecular gas. Because the birefringence is modulated at molecular revivals, it means that by adjusting the relative time delay between the pump and probe pulses, we can expect to obtain the probe's polarization state output as we desired. It would open up new possibilities for ultrafast information processing and telecommunications.

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