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Complete suppression of N₂ lasing by nonadiabatic molecular alignment effect in femtosecond filament

Yuxuan Zhang,^{1,*} Renjun Yang,^{2,3,*} Mingyang Zhuzou,¹ Aurélien Houard,⁴ André Mysyrowicz,⁴ Yi Liu,^{4,5} and Pengji Ding^{1,†}

¹*School of Nuclear Science and Technology, Lanzhou University, Lanzhou 730000, China*

²*Institute of High Energy Physics, Chinese Academy of Sciences, Beijing 100049, China*

³*Spallation Neutron Source Science Center, Dongguan 523803, China*

⁴*Laboratoire d'Optique Appliquée, ENSTA Paris, CNRS,*

Ecole polytechnique, Institut Polytechnique de Paris,

1024 Bd des Maréchaux, 91162 Palaiseau, France

⁵*Shanghai Key Laboratory of Modern Optical System,*

University of Shanghai for Science and Technology, 516, Jungong Road, Shanghai 200093, China

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The nonadiabatic molecular alignment effect is commonly found helpful in enhancing nonlinear processes such as high-order harmonics and strong-field ionization. In this work, an opposite phenomenon is observed, in which the nonadiabatic molecular alignment effect prepared with a linear-polarized prepulse strongly suppresses the generation of N₂ lasing induced by circularly-polarized 800 nm femtosecond laser pulse in filament plasma. The presence of a weak prepulse periodically suppresses the lasing at each revival timing of the rotational wave packet of the N₂ molecule. The underlying mechanism of the lasing signal suppression is attributed to sudden change of the polarization ellipticity of the pump laser pulse. Theoretical simulations by numerically solving the time-dependent Schrödinger equation with the molecular alignment effect included confirm sensitive changes of the polarization ellipticity of the pump laser at every alignment revival, which largely supports our interpretation.

I. INTRODUCTION

“Air lasing” phenomenon, which refers to the occurrence of a laser-like coherent emission from ultrashort laser-induced filamentary plasma in ambient air, has drawn considerable attention in recent ten years because of its rich physics in nonlinear and quantum optics on one hand, and potential applications in standoff detection on the other hand [1]. To date, such single-pass gain-swept lasing emission can be generated through three methods: (1) atomic lasing from oxygen/nitrogen species induced by deep-ultraviolet (UV) two-photon resonant pumping after predissociation of air molecules [2–5], or argon lasing using 3-photon resonant pumping [6]; (2) molecular lasing from neutral nitrogen (N₂) induced by circularly-polarized femtosecond laser pulse when it undergoes filamentation [7–9]; (3) molecular lasing from singly-ionized nitrogen N₂⁺ generated in femtosecond filament by linearly-polarized femtosecond laser pulse [10–17]. Deep-UV pump laser suffers from strong absorption and scattering when it propagates in ambient air, hence hindering the generation and practical application of atomic lasing in atmospheric remote sensing. As for N₂⁺ lasing, although weak optical gain at 391 nm wavelength was experimentally achieved in the backward direction [18], it mainly occurs in the forward direction. Therefore, near-infrared laser-induced N₂ lasing becomes

the most promising candidate for the laser source of atmospheric standoff detection in the future.

N₂ lasing occurs at 337 nm wavelength, which corresponds to the transition between the third and second excited triplet states of neutral nitrogen molecules, i.e. C³Π_u⁺(ν = 0) → B³Π_g⁺(ν' = 0) (ν and ν' represent the vibrational quantum number) [19]. After strong-field ionization, femtosecond laser pulse with circular polarization produces free electrons that distribute at higher kinetic energies but with smaller number compared to using linearly-polarized pulse [20, 21]. The kinetic energies of electrons are high enough to exceed the threshold of electron impact excitation of X¹Σ_g⁺ → C³Π_u⁺ via inelastic electron-molecule collision. Since electron impact excitation of X¹Σ_g⁺ → C³Π_u⁺ has a higher cross section and lower threshold of electron's kinetic energy compared to X¹Σ_g⁺ → B³Π_g⁺ [22], more N₂ molecules are excited to C³Π_u⁺ compared to B³Π_g⁺. Hence, an inversion can be established between these two states, leading to 337 nm lasing emission.

So far, generating measurable N₂ lasing emission in ambient air is still difficult due to the strong quenching effect of oxygen molecules [7]. Therefore, it is vital to seek approaches for enhancing the lasing intensity above the detection limit. One commonly used approach takes advantage of the molecular alignment effect, which nonadiabatically aligns the molecules along certain directions using a weak prepulse in advance and then irradiates the transiently aligned molecules with a strong pump pulse. In such a dynamic alignment process, the interaction between the ultrashort prepulse with respect to the rotational periods of the molecule and the

* These authors contributed equally to this work.

† dingpj@lzu.edu.cn

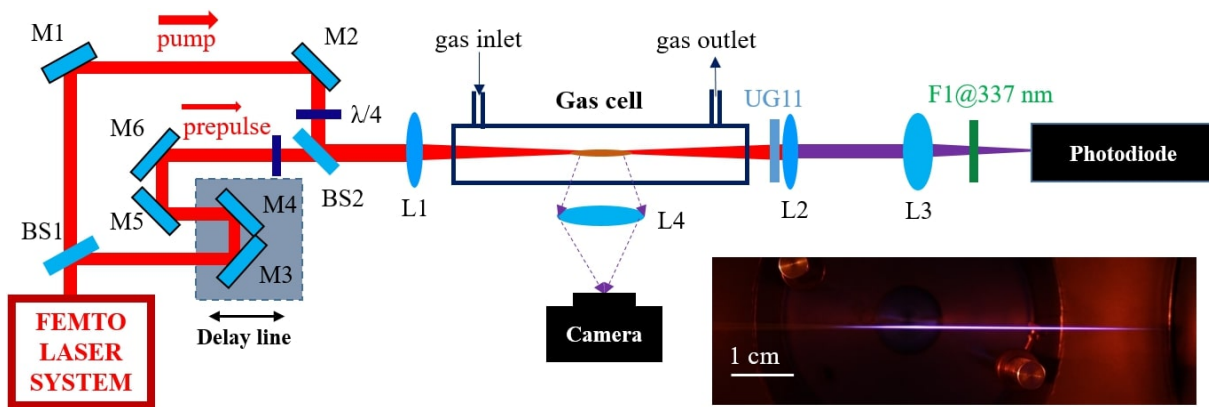


FIG. 1. Schematic of the experimental setup. BS: beamsplitter; M: high-reflectivity mirror; L: spherical lens; F: bandpass filter; $\lambda/4$: quarter waveplate. The time delay line (TDL) is formed by a pair of mirrors fixed on an electric-controlled one-dimensional translation stage. The inset shows a luminescence image of the filament plasma created in pure nitrogen.

molecular dipole moment gives rise to the excitation of a broad rotational wavepacket, whose coherence ensures alignment upon turnoff of the prepulse [23, 24]. Ultra-short laser-induced molecular alignment effect has been applied for manipulation or yield enhancement of high-order harmonic generation [25–27], strong-field ionization [28, 29], filament-forming laser propagation [30–32], N_2^+ lasing [33–35], etc., proving to be a useful approach. For instance, Litvinyuk *et al.* determined experimentally that the ionization probability of N_2 molecules aligned parallel to the laser field is four times higher than aligned perpendicular to it [28]. More than 50% enhancement of N_2^+ lasing intensity can be achieved at half-revivals of the rotational wave-packet created by the filament-forming laser pulse in nitrogen [34].

The impulsive molecular alignment effect also creates birefringence in the wake of laser filamentation. Béjot *et al.* demonstrated that the filament created with femtosecond laser pulse in atomic gases can induce substantial birefringence, which therefore acts like a wave plate that rotates the linear polarization of a delayed co-propagating probe pulse [36]. The transient birefringence originates from the difference between the nonlinear refractive indices induced by the instantaneous electronic response during laser filamentation on the axes parallel and orthogonal to its polarization. Yuan *et al.* quantified such ultrafast birefringence in argon by measuring the polarization rotation of a second-harmonic-generation probe pulse propagating through the filament [37]. It was also found that the polarization of a weak probe pulse can also be modulated near the field-free revivals of pre-excited rotational wavepackets created by 800 nm filamenting pulse [38]. The results suggest that the molecular alignment effect impulsively generates delayed birefringences in the wake of laser filamentation.

In this work, we investigate the influence of the non-adiabatic molecular alignment effect using an impulsive ultrashort laser on N_2 lasing in a prepulse-pump scheme. Unexpectedly, the introduction of the molecular align-

ment effect was found to drastically suppress the 337 nm lasing signal at both the full and fractional revivals of the rotational period of N_2 molecule.

II. EXPERIMENTAL METHOD

The experiments were carried out using a commercial Chirped Pulses Amplification (CPA) laser system (Thales Laser, model: Alpha 100). This system delivers femtosecond laser pulses at central wavelength of 800 nm with a duration of 45 fs and a maximum pulse energy of 15 mJ at a repetition rate of 100 Hz. Figure 1 schematically illustrates the experimental setup. The output laser beam was first separated into two arms by a beamsplitter (BS1), which reflects $\sim 10\%$ of incident laser energy for the prepulse arm. The prepulse passes through a time delay line to adjust the relative delay between the pump and prepulse. The pump laser passes through a quarter waveplate, by rotating which we can control its polarization. The pump and prepulse beam are combined with another beamsplitter (BS2) and then focused with the same spherical lens ($L1$, $f=1000$ mm) into a gas chamber filled with 1 bar pure nitrogen.

The circularly polarized pump laser pulse produces a bright filament plasma and generates 337 nm N_2 lasing in both the forward and backward directions. The lasing threshold is approximately 5.0 mJ. In this study, we only focus on the forward one. The forward lasing beam was collimated using a $f = 1000$ mm lens ($L2$) and then focused using a $f = 100$ mm lens ($L3$) into a calibrated photodiode. A UG11 filter, a BG39 filter, and a bandpass filter centered at 337 nm were used to block all interference light. The photodiode was connected to a lock-in amplifier and a digital acquisition system that controls the time delay line. The 337 nm lasing pulse energy was recorded in unit of mV and averaged for 100 shots for each delay time. By recording the lasing signal while scanning the relative delay between the pump

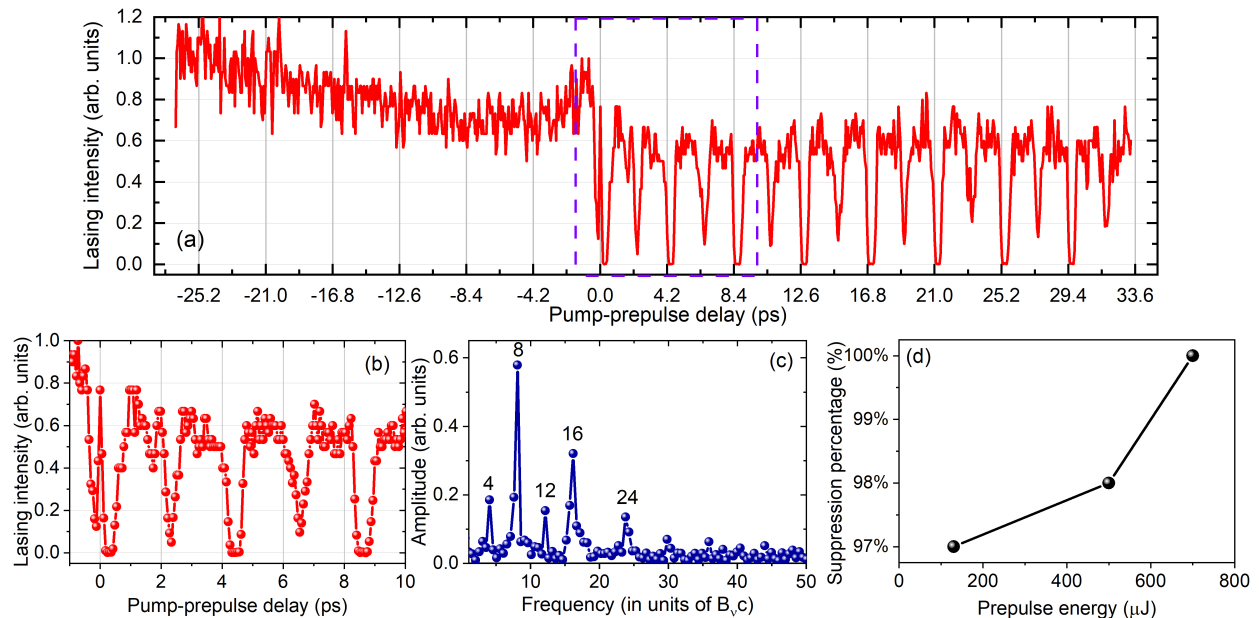


FIG. 2. (a) 337 nm lasing signal intensity as a function of relative delay between prepulse and pump pulses. The prepulse pulse energy was approximately $700 \mu\text{J}$. (b) Zoomed view of the result in (a) within the first cycle of the molecular revival of the N_2 molecule. (c) Corresponding FFT result of the result in (a) for delays larger than 0, where the frequency was represented in the unit of Bc . (d) Suppression percentage of the 337 nm lasing signal at half-revival versus the pre-pulse energy.

and prepulse, the influence of the nonadiabatic molecular alignment effect induced by the prepulse on the lasing generation can be then investigated. The scanning step of the delay was kept at 60 fs. During the experiments, the pump pulse energy was kept around 5.8 mJ while the prepulse energy was adjustable from 100 to $700 \mu\text{J}$.

III. RESULTS AND DISCUSSION

Figure 2 shows the 337 nm lasing signal as a function of the relative delay between the prepulse and pump pulse. The lasing signal is normalized with respect to that without the presence of the prepulse. Positive delay ($dt > 0$) means that the prepulse arrives before the pump pulse and vice versa. The zero delay was defined as that when the strongest interference pattern occurs, which is coincident with the appearance of a narrow peak shown in Fig. 2(b).

First, let's focus on the regime of $dt < 0$. When the prepulse laggies behind the pump pulse for more than ~ 25 ps, it has no noticeable influence on the lasing generation. As the prepulse approaches to the pump pulse, one can clearly see a gradual reduction of the lasing signal until $dt \sim -5$ ps. The lasing signal intensity quickly recovers within a few picoseconds as the prepulse further approaches the pump pulse. Just before the zero delay, ~ 1 ps, the lasing signal intensity reaches the local maximum, after which it sharply drops. Similar phenomena were previously observed in the pump-probe measurements of side fluorescence [39] and N_2^+ lasing [34, 35] from laser fil-

ament, but no clear explanation. Here, we interpret such behavior of 337 nm lasing signal upon changing the relative delay between the pump and prepulse for $dt < 0$ as a result of destructive interference of the lagged prepulse on the gain dynamics of N_2 lasing. The prepulse interrupts the evolution of gain by either promoting the molecules in the excited $\text{C}^3\Pi_u^+$ state to higher states or/and interacting with the plasma generated by the pump pulse. Equivalently, as the prepulse approaches the pump pulse from behind, it oppositely scans the profile of the gain dynamics of N_2 lasing. Indeed, we can see that the evolution profile and temporal scale of the lasing signal versus the delay for $dt < 0$ matches well with that of the gain dynamics previously reported in [21].

At zero delay, as clearly exhibited by the narrow peak in Fig. 2(b), no enhancement but approximately 20% reduction of the lasing signal was recorded. This can be attributed to field interference between the circularly polarized pump pulse and the linearly polarized prepulse. For $dt > 0$, on the basis of a general reduction of $\sim 40\%$, the lasing signal experiences periodic reductions at the revival times of the rotational wave packet of N_2 that has been pre-created by the prepulse. The general reduction of the lasing signal lasts more than 35 ps, which can be attributed to the presence of weak plasma generated by the prepulse. This attribution is verified by the fact that a weaker prepulse alleviates the general reduction. Surprisingly, the periodic reduction occurs at every revival time, suggesting that the molecular alignment effect generally suppresses the 337 nm lasing generation.

We repeated the measurements with another two pre-

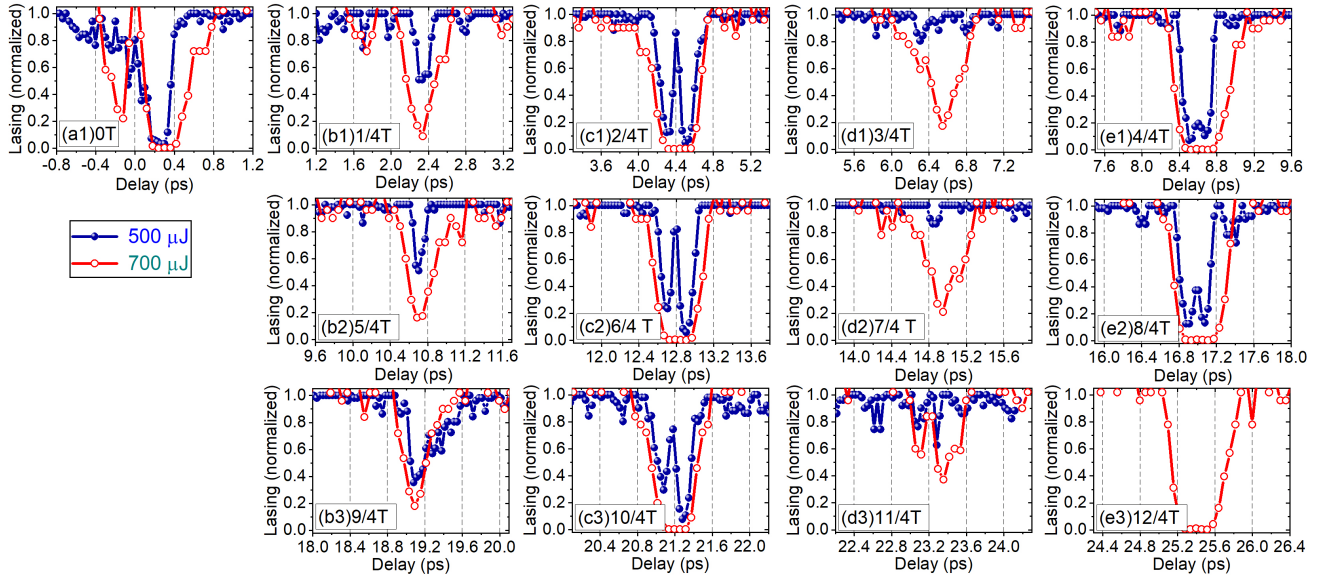


FIG. 3. Fine structures of the normalized lasing signal versus the pump-prepulse delay for two prepulse energies of $500 \mu\text{J}$ and $700 \mu\text{J}$ in the vicinity of (a) zero time delay, (b) $(n + 1/4)T$, (c) $(n + 2/4)T$, (d) $(n + 3/4)T$, and (e) $(n + 4/4)T$ alignment revivals.

pulse energies, e.g. 140 and $500 \mu\text{J}$. As shown in Fig. 2(d), a much weaker prepulse of $140 \mu\text{J}$ can still lead to 97% suppression of the lasing signal at half-revivals compared to that without the prepulse. This result exhibits a clear difference from that of side fluorescence [39] and N_2^+ lasing [34, 35], in which the recorded signals become maximum when the molecules are aligned along the polarization direction of the prepulse and becomes minimum when they are anti-aligned.

Figure 3 shows detailed comparison between the results of two different prepulse energies around a series of alignment revivals including $(n + 1/4)T$, $(n + 2/4)T$, $(n + 3/4)T$ and $(n + 4/4)T$, where $n = 0, 1, 2$ represents integer numbers and T denotes the rotational period of N_2 molecule. From Fig. (a1) and (e1)-(e3), the rotational period can be determined as $T = 8.406$ ps. It is very close to the theoretical revival period of neutral nitrogen molecule in the ground state $X^1\Sigma_g^+(\nu = 0)$. Given the rotational constant in the equilibrium position $B_e = 1.99824 \text{ cm}^{-1}$ and the first-order ν -dependent rotational constant $\alpha_e = 0.017318 \text{ cm}^{-1}$, one can get the rotational constant for $X^1\Sigma_g^+(\nu = 0)$ state as $B_\nu = B_e - \alpha_e(\nu + 1/2) = 1.989581 \text{ cm}^{-1}$ [40], which corresponds to a full revival period $T' = 1/(2B_\nu c) = 8.382$ ps. The slight difference between the measured revival period and the calculated one may originate from two contributions: the centrifugal distortion effect that causes slightly longer rotational periods for higher rotational levels and the potential existence of small amount of molecular nitrogen isotopes including $^{15}\text{N}_2$ and $^{14}\text{N}^{15}\text{N}$ that have smaller rotational constants.

Several observations can be drawn from Fig. 3 as follows: (1) The first one is that the extents of lasing signal suppression at $(n + 1/4)T$ and $(n + 3/4)T$ revivals are less

than that at $(n + 2/4)T$ and $(n + 4/4)T$ revivals, which can be certainly attributed to their smaller alignment degrees. (2) The lasing signal suppression at $(n + 3/4)T$ revivals is apparently weaker than that at $(n + 1/4)T$ revivals for $500 \mu\text{J}$ prepulse case, which is however not that obvious for $700 \mu\text{J}$. (3) Compared to the results of $700 \mu\text{J}$ prepulse where the lasing signal was completely suppressed at $(n + 2/4)T$ and $(n + 4/4)T$, the results of $500 \mu\text{J}$ prepulse show prominent regenerations of the lasing signal at these two alignment revivals. (4) As clearly shown in Fig. 3(c) and (e), the lasing signal regeneration at $(n + 2/4)T$ revival is much more prominent than that at $(n + 4/4)T$. (5) The extent of the lasing generation at $(n + 2/4)T$ revival steadily decreases from approximately 0.90 at $(1/4)T$ to 0.75 at $(10/4)T$, whereas the results at $(n + 2/4)T$ revival show increasing lasing regeneration. (6) An asymmetry of two suppression maxima around the lasing regeneration peak can be well observed in Fig. 3(c1)-(c3). It becomes more significant as the time delay increases from $(2/4)T$ to $(10/4)T$, in which the right suppression maxima seem to remain unchanged but the left maxima steadily decrease. Such asymmetry behaves just the opposite for $(n + 4/4)T$ if one looks into the results in Fig. 3(e1) and (e2).

As a direct comparison, we performed another prepulse-pump experiment using a circularly polarized prepulse with an energy of $\sim 730 \mu\text{J}$. The dependence of 337 nm lasing signal on the relative delay is shown in Fig. 4. Note that a circularly polarized femtosecond laser pulse can also produce the molecular alignment effect [41, 42], but it occurs in the propagation direction and hence does not affect the pump laser in the cross-section plane. As expected, the 337 nm lasing signal is dramatically increased as two pulses spatiotemporally

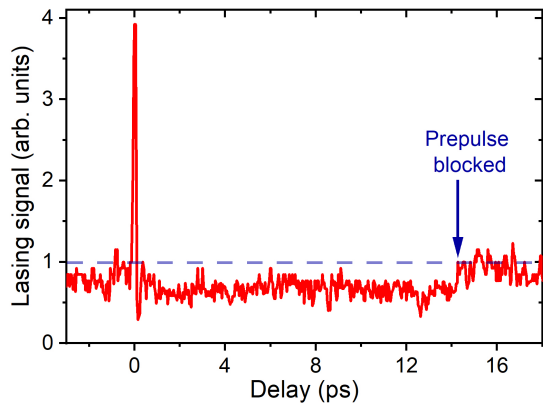


FIG. 4. 337 nm lasing signal intensity as a function of the relative delay between the circular pump pulse and circular prepulse. The timing when the prepulse was blocked intentionally is denoted.

overlapped, an outcome of laser field interference. A weak suppression of the lasing signal is noticeable, which can be ascribed to the presence of weak plasma induced by the prepulse. Once the prepulse is artificially blocked, as indicated by the arrow in Fig. 4, the lasing signal intensity recovers.

The molecular alignment effect was found to be able to induce ultrafast birefringence in isotropic gas, which can change the polarization state of a delayed laser pulse [36–38, 43–45]. Meanwhile, we have shown in our previous studies [7, 8] that the generation of N_2 lasing in femtosecond laser filamentation only occurs when the polarization state of the pump laser pulse is very close to being circular. With the pump pulse energy of approximately 5.8 mJ, we measured the dependence of the lasing signal on the polarization ellipticity of the pump laser, as shown in Fig. 5. Same as usual, the zero and 90 degrees of the QWP rotation angle correspond to linear polarization whereas 45 degrees corresponds to circular polarization. One can see that the lasing signal rapidly decreases to the fluorescence level as the polarization was changed slightly away from circular. To estimate how quickly the signal decreases, a Gaussian function was applied to fit the data in Fig. 5, and the FWHM of the dependence curve is about $\Delta\epsilon = 0.3$. It indicates that the signal drops by 50% if the polarization ellipticity is changed by only 0.15 from circular.

By combining the above information, we speculate that the origin of lasing signal suppression at different revivals in Fig. 3 is that the ultrafast birefringence induced by the linearly-polarized prepulse changes the polarization of the energetic pump pulse from initial circular and thus leads to significant reduction of the lasing signal. To confirm our hypothesis and better understand the experimental results, we simulated the polarization ellipticity dynamics of the pump laser pulse in the presence of a weak prepulse. We denote that the prepulse polarizes along the x-axis and propagates along the z-axis. The

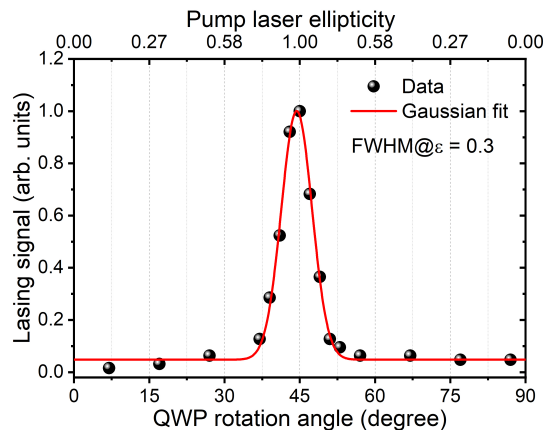


FIG. 5. Normalized 337 nm lasing signal versus the rotational angle of $1/4\text{-}\lambda$ waveplate. Gaussian fit was applied to the data to extract the FWHM of the dependence curve.

electric field of the circularly polarized pump pulse can be combined by sinusoidal oscillations $E_x(t, z)$ and $E_y(t, z)$, respectively. Their amplitudes E_{x0} and E_{y0} are equal with an phase difference $\delta_0 = \delta_{x0} - \delta_{y0} = \pi/2$. Due to the ultrafast birefringence effect, the refractive index would be modulated when the prepulse interacts with the gas medium, which leads to an external phase difference between $E_x(t, z)$ and $E_y(t, z)$ and hence change the ellipticity of the pump pulse. The ellipticity can be described by $\tan(\delta/2)$, where δ is limited to the range of $0 \leq \delta \leq \pi/2$.

In molecular gas medium, the refractive index modulation does not only originate from the electronic Kerr effect but also from plasma formation and molecular alignment. The refractive index difference induced by the first contribution is proportional to the prepulse intensity $I_{pre}(t)$ and the non-linear refractive index n_2 [46]:

$$\begin{aligned} \Delta n_{Kerr,xy}(t) &= \Delta n_{Kerr,x}(t) - \Delta n_{Kerr,y}(t) \\ &= \frac{4}{3} n_2 I_{pre}(t). \end{aligned} \quad (1)$$

The generation of a filament plasma reduces the local refraction index according to the law [47]

$$\Delta n_{plasma}(t) = -\frac{\rho_e(t)}{2\rho_c}, \quad (2)$$

where ρ_e is the density of free electrons and $\rho_c = \epsilon_0 m_e \omega_0^2 / e^2$ denotes the critical plasma density above which the plasma becomes opaque. As usual, ϵ_0 , m_e and e denote the vacuum permittivity, electron mass and charge, respectively. For a driving laser wavelength of 800 nm, $\rho_c \sim 1.7 \times 10^{21} \text{ cm}^{-3}$. In filaments, the widely reported value of ρ_e was on the order of 10^{16} cm^{-3} [48], which then results in $\Delta n_{plasma} \sim -3 \times 10^{-6}$. Besides, it was found that N_2 molecules are four times more likely to ionize when aligned parallel to the laser field than when aligned perpendicular to it [28]. Hence it is reasonable to speculate that the electron density distribution follows

the same ratio, e.g. $\rho_{e,x} \approx 4\rho_{e,y}$ for x-axis polarized laser pulse. It then leads to an estimation of negative refractive index difference $\Delta n_{plasma,xy} \sim -2 \times 10^{-6}$. Since the electron-ion recombination is usually of the order of nanoseconds, the filament can be considered as a stationary plasma within the probed delay range of 35 ps and hence the negative contribution of plasma formation to the refractive index difference stays constant.

The time-dependent refractive index difference caused by the nonadiabatic molecular alignment is given by [46]:

$$\begin{aligned} \Delta n_{align,xy}(t) &= \Delta n_{align,x}(t) - \Delta n_{align,y}(t) \\ &= 3 \frac{\pi N}{n_0} \Delta \alpha \left(\langle \langle \cos^2 \theta \rangle \rangle (t) - \frac{1}{3} \right), \end{aligned} \quad (3)$$

where N and n_0 respectively represent the medium density and the refractive index upon the weak field, and $\Delta \alpha = \alpha_{\parallel} - \alpha_{\perp}$ is the polarizability anisotropy defined by the difference between the polarizability components perpendicular and parallel to the molecular axis. The degree of molecular alignment is represented by $\langle \langle \cos^2 \theta \rangle \rangle (t)$, which can be obtained by following steps [49]. As a linear molecule, the rotational wave packet of N_2 after ultrashort excitation can be expanded by the rotational eigen states $|JM\rangle$:

$$\psi(t) = \sum_J c_J(t) e^{-i \frac{E_J}{\hbar} t} |JM\rangle, \quad (4)$$

where $c_J(t)$ is the complex time-dependent coefficient, and $E_J = \hbar c B J(J+1) - \hbar c D J^2(J+1)^2$ is the eigen energy of rotational state with the rotational Hamiltonian H_{rot} . Here, B and D are rigid and nonrigid rotor constants respectively. The evolution of $\psi(t)$ was obtained by solving the time-dependent Schrödinger equation (TDSE):

$$i\hbar \frac{\partial}{\partial t} \psi(t) = [H_{rot} - \pi \varepsilon_0 F^2(t) (\Delta \alpha \cos^2 \theta + \alpha_{\perp})] \psi(t), \quad (5)$$

where ε_0 and $F(t)$ are the vacuum permittivity and the electric field of the laser pulse. After obtaining the time-dependent coefficient $c_J(t)$ by Eq. 5, the alignment degree of an initial eigen state $|JM\rangle$ can be calculated:

$$\langle \cos^2 \theta \rangle_{JM}(t) = \langle \psi_{JM}(t) | \cos^2 \theta | \psi_{JM}(t) \rangle, \quad (6)$$

where the initial state of $\psi_{JM}(t)$ is $|JM\rangle$. By considering the Boltzmann distribution of the initial molecular states, the alignment degree of the molecules can be calculated:

$$\langle \langle \cos^2 \theta \rangle \rangle (t) = \sum_J W_J \sum_{M=-J}^J \langle \cos^2 \theta \rangle_{JM}(t), \quad (7)$$

where W_J is the Boltzmann weight factor. After calculating the refractive index difference induced by the prepulse, the phase difference of the pump pulse between the x -axis and the y -axis can be obtained by:

$$\begin{aligned} \delta(t) &= \frac{2\pi}{\lambda} [\Delta n_{Kerr,xy}(t) + \Delta n_{plasma,xy} \\ &\quad + \Delta n_{align,xy}(t)] L + \delta_0, \end{aligned} \quad (8)$$

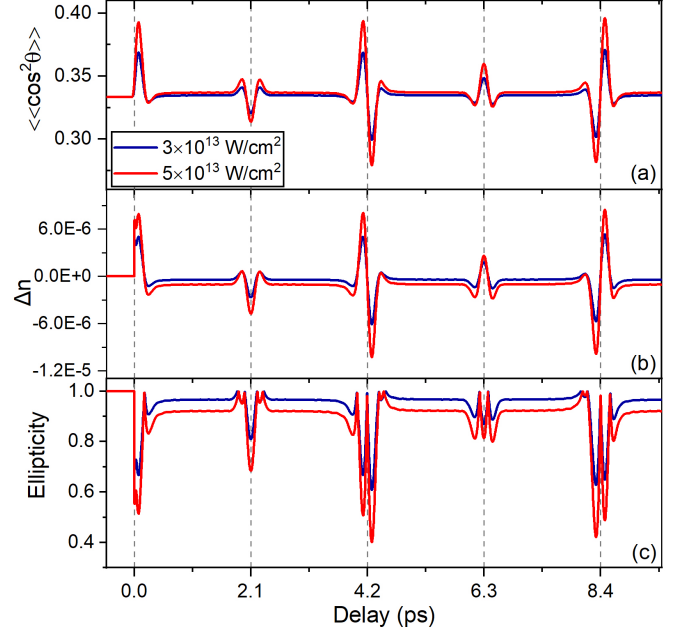


FIG. 6. Simulated results of (a) the molecular alignment degree $\langle \langle \cos^2 \theta \rangle \rangle$, (b) the refractive index change and (c) the polarization ellipticity of the pump laser pulse as a function of the relative time delay between the circular pump laser pulse and the linear prepulse.

where λ is the center wavelength of the pump pulse.

By assuming that the prepulse mainly interacts with the nitrogen gas in a cylindrical filamentary column, we numerically simulated the modulation of the polarization ellipticity of the pump pulse induced by a prepulse with two different laser intensities, $3 \times 10^{13} \text{ W/cm}^2$ and $5 \times 10^{13} \text{ W/cm}^2$. For these two prepulse intensities, we set $\Delta n_{plasma,xy}$ to be -6×10^{-7} and -2×10^{-6} , respectively. The filament length was set to $L = 1 \text{ cm}$. Figure 6 shows simulated evolution of the molecular alignment degree $\langle \langle \cos^2 \theta \rangle \rangle$, the total refractive index change Δn and the polarization ellipticity of the pump laser as the relative delay between the circular pump and linear prepulse varies within the first revival period. Most importantly, one can observe that the polarization ellipticity of the pump laser will be significantly modulated in the wake of the linear prepulse around the vicinity of integral multiples of the quarter revival, in which stronger alignment effect results in larger ellipticity shift from initial circular polarization. If using an intenser prepulse, the ellipticity modulation of the pump pulse becomes more significant. Given the generation of the 337 nm lasing is sensitive to the ellipticity of the pump pulse, a sudden polarization change of the pump laser pulse leads to the disappearance of the lasing signal when the ellipticity is lower than the threshold of approximately 0.7. Around the zero time delay, all three mechanisms including the instantaneous electronic Kerr effect, plasma formation and adiabatic molecular alignment effect contribute to the fast drop of

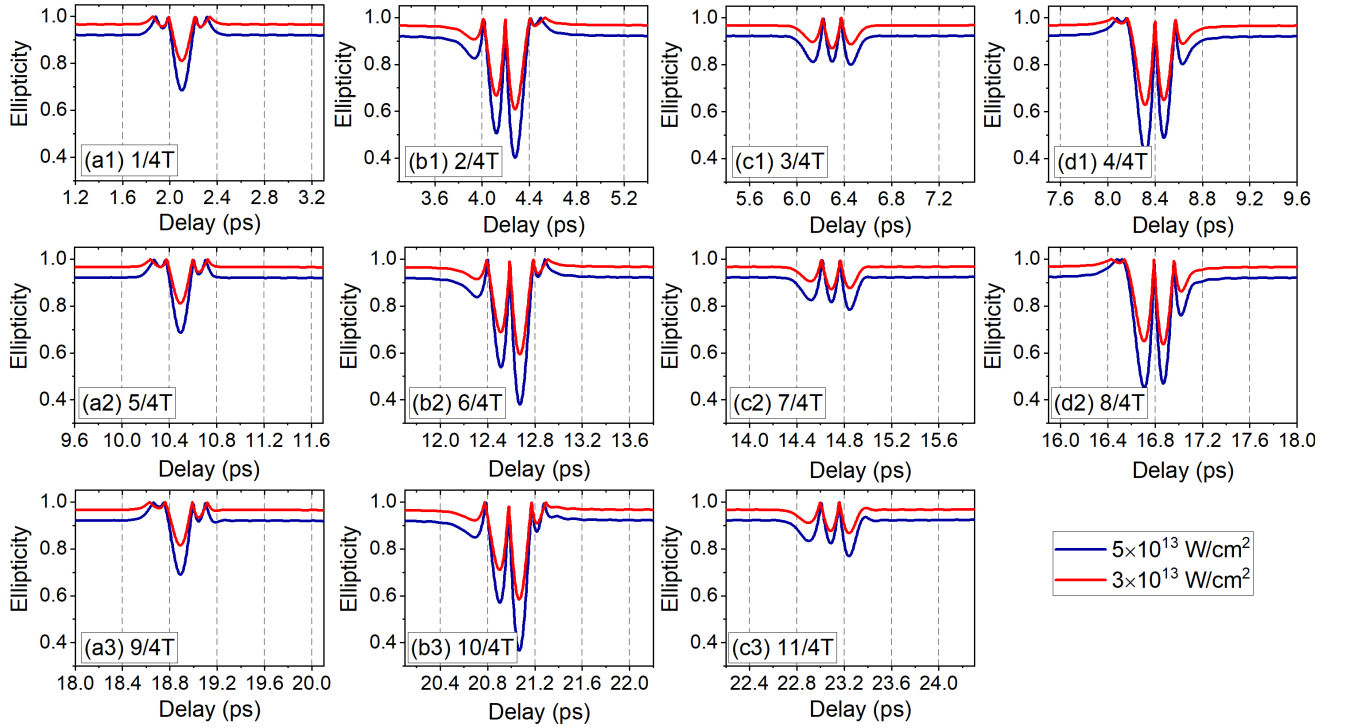


FIG. 7. Simulated polarization ellipticity of the driving laser pulse as a function of the relative time delay between the circular pump laser pulse and the linear prepulse in the revival vicinity of (a) $(n+1/4)T$, (b) $(n+2/4)T$, (c) $(n+3/4)T$, (d) $(n+4/4)T$ for two prepulse intensities: $3 \times 10^{13} \text{ W/cm}^2$ and $5 \times 10^{13} \text{ W/cm}^2$.

the pump laser ellipticity from $\epsilon = 1$. After the passage of the prepulse, the long-lived presence of plasma introduces a constant shift to the pump laser ellipticity whereas the nonadiabatic alignment effect leads to its periodic modulations. Note that the refractive index change due to molecular alignment effect is on the same order of magnitude with that owing to plasma formation, which manifests itself as the baseline in Fig. 6(b).

The simulated fine structure of the laser ellipticity evolution is shown in Fig. 7, which reproduces most of the features observed in the experiments. Firstly, the ellipticity shift at $(n+1/4)T$ exhibits more prominent than $(n+3/4)T$, which qualitatively agrees with the experimental feature with $500 \mu\text{J}$ prepulse shown in Fig. 3. Secondly, the ellipticity shift occurs at both aligned and anti-aligned moments, and the shift at the latter moment is obviously larger than that at the former one. If we compare the simulated results at $(n+2/4)T$ and $(n+4/4)T$ in Fig. 7(b) and (d), they show opposite characteristics that qualitatively agree with the experimental observations in Fig. 3(c) and (d). It should be noted that above two features can only be achieved after introducing the negative refractive index change from anisotropic plasma formation.

As the time delay increases, as shown in Fig. 7(b1)-(b3), the ellipticity shifts at the aligned moments around $(n+2/4)T$ revivals becomes smaller while the shifts at the anti-align moments increases, making the difference of

two peaks more significant. The simulated results around $(n+4/4)T$ revivals show exactly same trend. This feature is also in good agreement with the experimental observations in Fig. 3(c) and (d). The underlying reason of this time-dependent asymmetry between the ellipticity shifts at the aligned and anti-aligned moments relates to the detuning of the rotational wave packet induced by the centrifugal distortion effect that considers the nitrogen molecule as a nonrigid rotor. Higher rotational levels have slightly longer rotational periods and hence the temporal difference between these periods becomes increasingly larger with the time. That will lead to an increasingly larger asymmetry.

It should be noted that two features observed in the experimental results in Fig. 3, e.g. the damping recovery of the lasing signal with the time at $(n+2/4)T$ revival and a much stronger lasing recovery at $(n+2/4)T$ revival compared to $(n+4/4)T$, were not reproduced in the simulations. The first feature could originate from the collisional decay of rotational coherence, which was not included in the simulations. As a result of growing dispersion in the rotational wave packet with the time, the molecular alignment degrades and hence the induced birefringence effect for a delayed pump laser becomes weaker as the delay increases. The second feature certainly relates to the ultrafast transition direction of the rotationally-excited molecules from being aligned to anti-aligned or vice versa. The experimental results suggest

that the lasing recovery favors the transition direction from aligned to anti-aligned rather than the opposite. The failure to reproduce this feature in the simulation suggests that the current theoretical model is yet complete, which then requires deeper investigations.

IV. CONCLUSION

To conclude, we experimentally found that the molecular alignment effect plays a detrimental influence on 337 nm lasing generation of N_2 molecules in laser filamentation that is created by circularly polarized near-IR femtosecond laser pulse. The molecular alignment prepared by a linearly polarized near-IR prepulse, which is much weaker and temporally ahead of the pump laser pulse, periodically suppresses the lasing generation at each revival of rotational wave packets. A prepulse with only 140 μJ energy can suppress the lasing signal intensity by 97%. It was observed that stronger alignment degree leads to more significant lasing suppression. The fine evolution structures of the lasing signal near the revivals were analyzed, which show distinct features between the structures around $(n + 1/4)T$ and $(n + 3/4)T$ as well as the structures around $(n + 2/4)T$ and $(n + 4/4)T$. The lasing suppression at $(n + 1/4)T$ revival appears stronger than that at $(n + 3/4)T$ one. A growing asymmetry between two suppression peaks at $(n + 2/4)T$ revival with increasing time delays between the pump pulse and prepulse was observed whereas an opposite evolution was noticed at $(n + 4/4)T$ revival.

The underlying mechanism of the lasing suppression by the molecular alignment effect was hypothetically attributed to the sudden change of the polarization ellipticity of the pump laser pulse when it experiences ultrafast birefringence induced by the prepulse. Theoretical simulations by numerically solving the TDSE reveal sensitive changes in the polarization ellipticity of the pump laser pulse at every alignment revival. The simulated behavior of the pump laser ellipticity captured most of the observed features in the fine structure of lasing suppression, such as stronger alignment degree inducing a larger ellipticity drop and more significant asymmetry between two suppression peaks at $(n + 2/4)T$ with the increasing time delay. However, the simulation fails to reproduce two experimental observations, e.g. the damping recovery of the lasing signal at $(n + 2/4)T$ revival and a much stronger lasing recovery at $(n + 2/4)T$ revival compared to $(n + 4/4)T$. Possible reasons were briefly discussed. In the end, it becomes clear that current theoretical model requires further completeness to fully reproduce the experimental observations.

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