

Spectral-width variation of N_2^+ lasing

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The complicated collisional processes inside a femtosecond laser-induced plasma column can show a great impact on the light emissions from it. A known example is that the lifetime of noncoherent fluorescence emitting from gas plasma can be greatly decreased due to the fierce ultrafast plasma collisions. However, for coherent stimulated emissions in plasma surrounding, to what extent it can be influenced by the collisional process has not been fully understood. Herein, we report an experimental spectral width analysis on coherent N_2^+ lasing that is generated in weakly ionized gas plasma with a frequently adopted pump-probe scheme. It is found unexpectedly that the spectral width of N_2^+ lasing evidently shortens with the increasing delay between the pump and the probe pulses at a pressure of 40 mbars, whereas an invariable spectral width appears when the gas pressure is decreased below 10 mbars. Moreover, it was also observed that the spectral width narrows with the increment of polarization angle between the pump and the probe pulses. These observations update the understandings of molecular spectral manipulation on the basis of the previous that the spectral modulations mainly occur in the vicinity of molecular alignment. Our results indicate that by combining the spectral width and intensity analysis, a more comprehensive understanding regarding the impact of ultrafast electronic plasma-ion collisions on the N_2^+ lasing generation can be obtained. These findings also shed light on the spectral manipulation for coherent irradiations produced during strong laser-matter interaction.

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I. INTRODUCTION

Several lasing lines corresponding to the bound-state transitions of N_2^+ can be triggered when neutral nitrogen molecules at suitable pressures are subjected to an intense femtosecond laser field [1–10]. The phenomenon of N_2^+ lasing attracts continuous attention in recent years because of not only its great promise in remote atmospheric sensing, but also its elusive gain and amplification mechanisms, and stimulates many frontier investigations on strong-field quantum optics of molecular ions currently [11–14]. The two representative lasing lines are 391 nm and 428 nm, which correspond to the transitions from the electronic-vibrational states of $B^2 \Sigma_u^+(\nu = 0)$ to $X^2 \Sigma_g^+(\nu = 0)$ and $X^2 \Sigma_g^+(\nu = 1)$, respectively. The amplitude, polarization and phase information of the N_2^+ lasing have been widely investigated. It is suggested that the 391-nm and 428-nm lasing produced in a plasma column or a filament can last for several picoseconds and their intensities can be several orders of magnitude stronger than the injected one [4,13,15]. Besides, the spectral intensity of 391-nm lasing can be significantly enhanced by exploiting a polarization-tailored pump pulse [16–18] or coherently modulated by a dual-pulse excitation [19–21].

The readily observed ionic lasing by a typical pump-probe scheme shows many complicated facets due to the additional

involvement of molecular vibrational and rotational degrees. Although various theoretical models, including the stationary three-state coupling [22–24], dynamic strong-field ionization coupling [25], and electronic recollision models [4], have been proposed to interpret some of experimental observations taking place under a certain condition, such as population inversion [17,23,24], electronic and rotational quantum coherences [9,19–21], a clear unified picture has not been achieved regarding the mechanism of N_2^+ lasing so far. It is noteworthy that one of the key experimental parameters that obscures the creation process of ionic lasing is gas pressure, the amount of which usually could change the pump peak intensity at the focus, plasma density, and dephasing time, etc. Particularly, an essential relevant issue is that to what extent the decoherence or depopulation process of the N_2^+ system brought by electronic impact should be taken into consideration remains a challenge. It was reported that for a low gas pressure from a few to tens of mbars, the prepared electronic coherence of N_2^+ resulting in macroscopic quantum amplification [4,13,15,19–21], i.e., superradiance, was considered being immune to the collisional process. A similar study revealed that the electron-atom collisions in a weakly ionized plasma column were crucial for high harmonic spectroscopy [26].

In the present paper, we find direct evidence to show how the ultrafast collisions between electrons and ions influence the spectral width of N_2^+ lasing from the perspective of spectroscopy by which the underlying physical mechanism is clarified. By studying the relation of spectral width of N_2^+ lasing to the experimental parameters encompassing the gas

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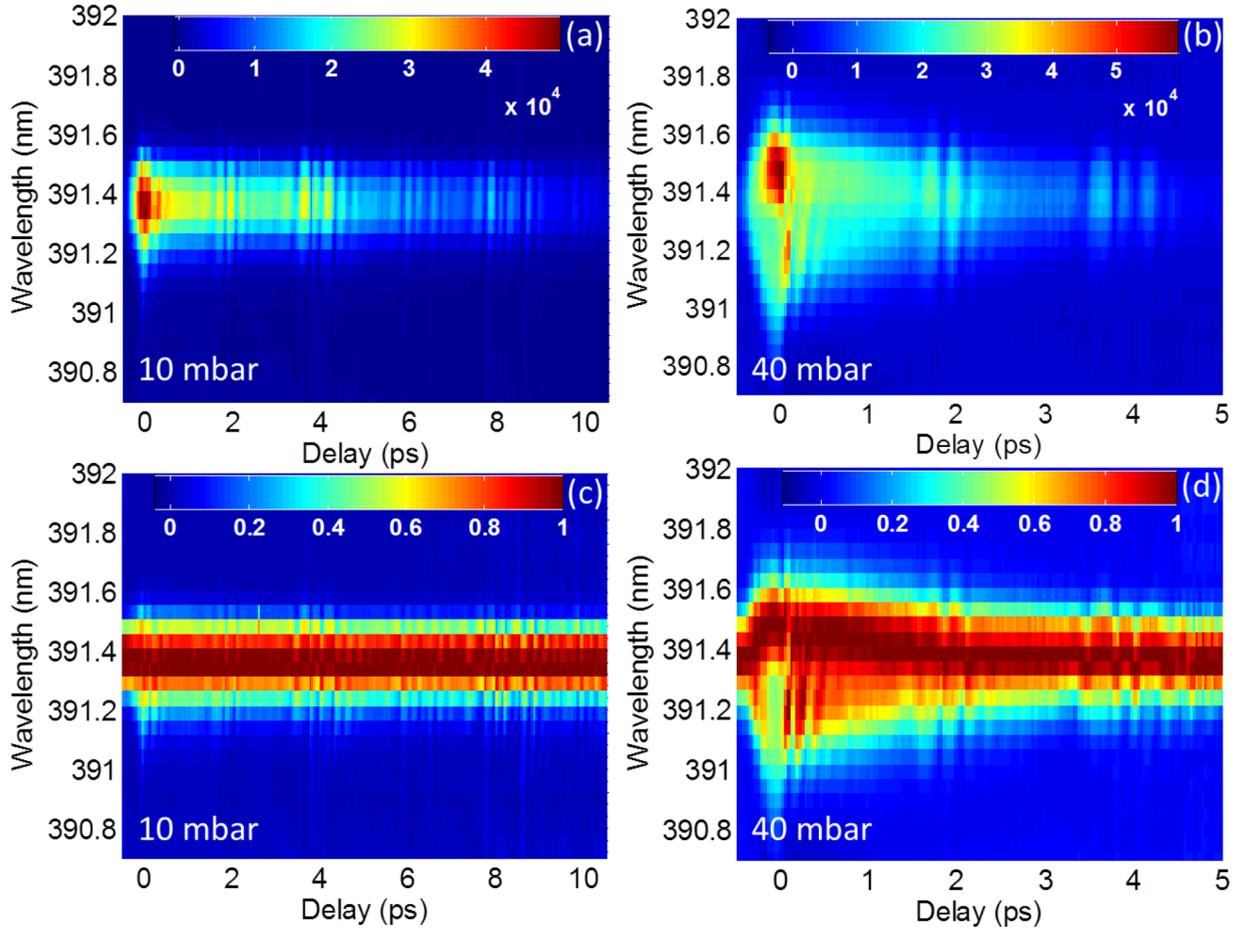


FIG. 1. The unnormalized N_2^+ lasing spectrum as a function of the pump-probe delay at a pressure of (a) 10 mbars and (b) 40 mbars. (c) and (d) same as (a) and (b), respectively, but the lasing spectrum at each pump-probe delay is normalized by their spectral peaks. Note that the delay axis range of the plots at 40 mbars excludes the last 5 ps due to the fast decay.

pressure, pump-probe delay, and polarization angle θ , our results clearly demonstrate that the electron-ion collisions can be important in varying spectral width at nonalignment delays by changing the gain lifetime of N_2^+ lasing, and, therefore, the decay of electronic coherence should be taken into consideration in the model of simulating N_2^+ lasing generation, even at a relatively low gas pressure, which has been overlooked in most prior instances.

II. EXPERIMENTAL SETUP

The pump-probe experimental setup is similar to that used in our previous works [3,9,15]. Briefly, intense femtosecond laser pulses (3.6 mJ/pulse, 35 fs, and 795 nm, 1 kHz) delivered from a commercial Ti:sapphire laser system were divided into two paths by a 7:3 beam splitter. The main pulse with the energy of 1.9 mJ acted as the pump. The other beam after being frequency doubled by a 0.2-mm-thick barium borate (BBO) crystal was used as the probe. A thin-film polarizer was placed after the BBO crystal to ensure the linear polarization characteristic of the probe pulse, and then a half-wave plate at 397 nm was installed to vary the probe polarization direction. The polarization direction of the pump with respect to that of the probe is defined as θ . The time interval between the pump and the probe was controlled by a motorized

translation stage with a resolution of ~ 670 as. Both beams were combined with a dichromatic mirror (high reflectivity at 800 nm and high transmission at 400 nm) and were collinearly focused with an $f = 30$ -cm lens into a gas chamber filled with pure nitrogen. The forward generated N_2^+ lasing signals after collimation by a $f = 25$ -cm lens were recorded with a collecting system consisting of an integrating sphere and a grating spectrometer (Kymera-328I, Andor), which can minimize the polarization-induced different responses to the detector. In our measurement, the width of the entrance slit of the spectrometer was fixed at $250 \mu\text{m}$.

III. RESULTS

Figures 1(a) and 1(b), respectively, show the recorded 391-nm N_2^+ lasing [the electronic transition can be assigned to be $B^2 \Sigma_u^+(\nu = 0) \rightarrow X^2 \Sigma_g^+(\nu = 0)$] spectrum as a function of the time delay between the pump and the probe pulses at the gas pressure of 10 mbars and 40 mbars. The polarization direction was set mutually parallel for the pump and probe in these cases, i.e., $\theta = 0^\circ$. It can be seen that the lasing intensity gradually decreases with the increasing pump-probe delay for both the pressures due to the weakened gain factor, which is consistent with our previous works [2,3]. Besides, at the pressure of 10 mbars, the lasing signal lasts for about

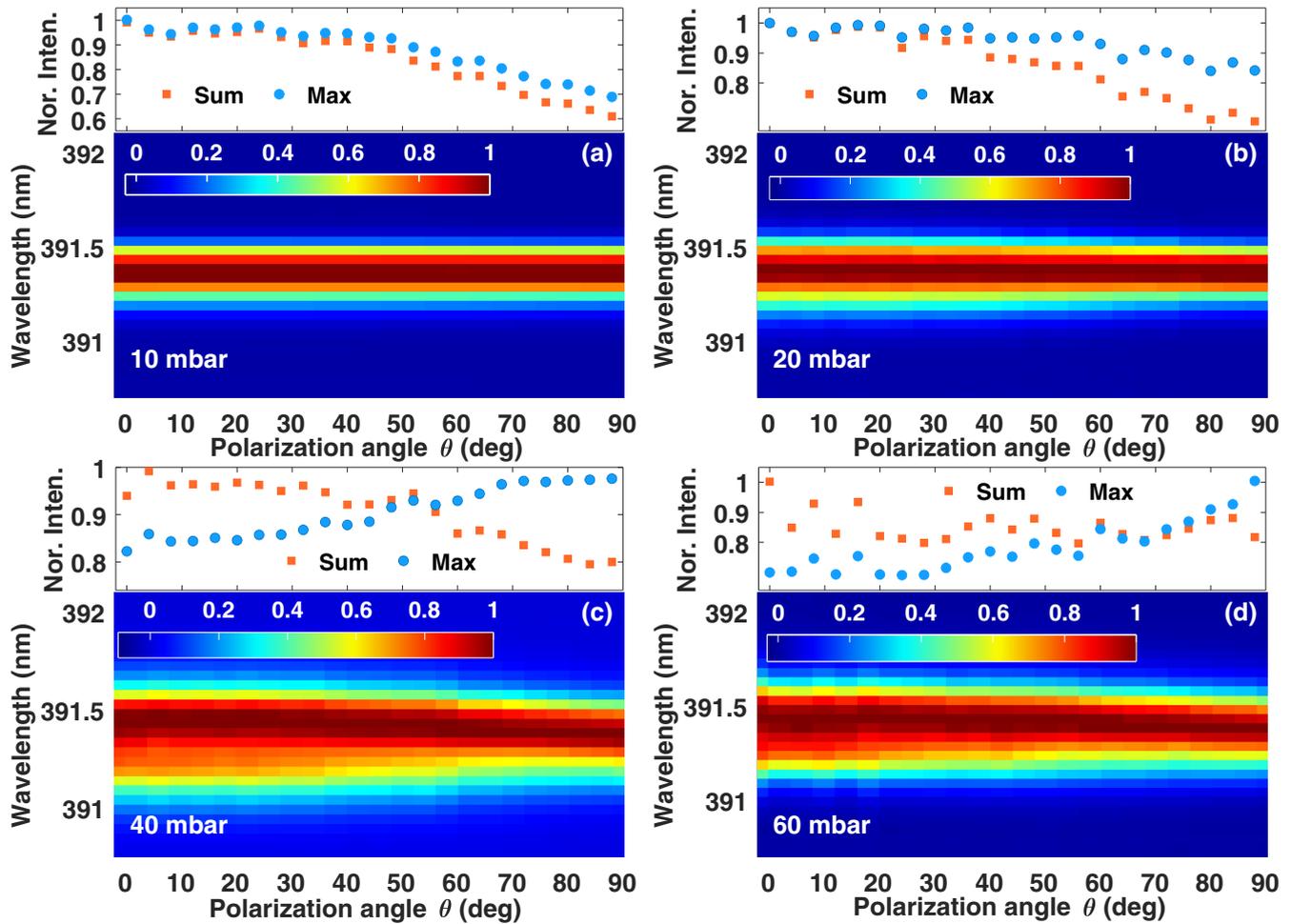


FIG. 2. The normalized N_2^+ lasing spectrum as a function of the polarization angle θ between the linearly polarized pump and the probe pulses at the pressure of (a) 10 mbars, (b) 20 mbars, (3) 40 mbars, and (d) 60 mbars. On the top of each panel, we also show the peak intensity and integrating intensity of N_2^+ lasing (summing over the spectral components from 390.6 to 392 nm) as a function of the polarization angle θ .

10 ps (from the moment of birth to that of disappearance) whereas as the pressure is increased to 40 mbars, the lasing signal only survives within around 5 ps. Another interesting observation is that the spectral width obviously shortens with the increment of the pump-probe time interval when the gas pressure was fixed at 40 mbars, which is distinctly different with that at the pressure of 10 mbars. In order to better analyze the peculiar spectral width variation, we perform normalization for the measured spectral intensity at every pump-probe instants by its peak intensity. The corresponding results are presented in Figs. 1(c) and 1(d), separately. It can be clearly seen that for the case of 10 mbars, the spectral width basically keeps invariable with the pump-probe delay whereas for a relative high pressure of 40 mbars, the spectral width gradually decays with the increasing delay. It deserves mentioning that the similar spectral width narrowing effect was observed at an ambient pressure based on a relatively loose focusing configuration [6]. However, the variation tendency of the spectral width versus the pump-probe delay exhibit a big difference with ours due to the difference of experimental conditions. Additionally, we also noted the apparent spectral modulation of the N_2^+ lasing around 2 and 4 ps due to the instantaneous phase control caused by molecular align-

ment of ions, which has been reported in several prior works [2,3,5,6].

Figure 2 shows the dependence of the measured N_2^+ lasing spectra on the polarization angle θ at various gas pressures. The spectra were normalized by its peak spectral intensity for each measurement. The delay between the pump and probe pulses was fixed at a nonalignment delay of ~ 0.8 ps in these measurements. Of note, the nonalignment delay herein solely refers to the moment when the contribution of revivals of a molecular rotational wave packet to molecular alignment is negligible. But the preferential ionization and anisotropic resonant electronic couplings generated within the pump duration can give rise to a certain permanent alignment of molecules, which can show some impact on the subsequent creation of N_2^+ lasing. It can be seen that the spectral width of the ionic lasing slightly narrows with the increment of the polarization angle θ for the pressures of 20, 40, and 60 mbars, and the tendency becomes more evident as the pressure grows, whereas the spectral width exhibits rare changes at a relatively low pressure of 10 mbars. The spectral width variation mechanism for the observations will be discussed later. Additionally, another interesting observation can be obtained from the top of the each panel is that the peak intensity of the N_2^+ lasing

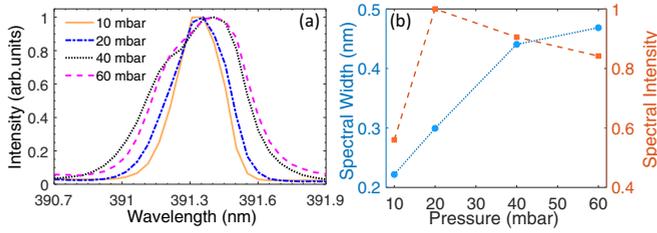


FIG. 3. (a) The typical normalized N_2^+ lasing spectra at several pressures for the parallel configuration, i.e., $\theta = 0^\circ$. (b) The dependence of spectral width of full width at half maximum (FWHM) on the gas pressure by interpolating the spectra in (a). Meanwhile, the lasing intensity by integrating the spectral components from 390.6 to 392 nm versus the gas pressure is also plotted.

spectrum decreases with the increasing polarization angle θ for the pressures of 10 and 20 mbars, which, however, is reversed for the pressures of 40 and 60 mbars. As can be seen from Figs. 2(c) and 2(d), the spectral width of N_2^+ lasing obviously decreases with the increasing polarization angle θ at these two pressures. The maximum spectral intensity at $\theta = 0^\circ$ can be smaller than that at $\theta = 90^\circ$, but due to the broader band width for the case of $\theta = 0^\circ$, the spectral intensity is larger than that at $\theta = 90^\circ$ when adopting the spectral integrating method. Therefore, the existing spectral width variation phenomenon in N_2^+ lasing could lead to totally different tendencies for studying the dependence of lasing intensity on the angle θ when processing the experimental data using spectral integrating or the peak spectral intensity.

Figure 3(a) shows the separately normalized N_2^+ lasing spectra at the pressures of 10, 20, 40, and 60 mbars for the parallel polarization configuration (i.e., $\theta = 0^\circ$). It can be clearly seen that the spectral width obviously increases with the growing pressure. With the interpolation fitting method, the quantitative spectral widths of FWHM in Fig. 3(a) are obtained as displayed in Fig. 3(b). Meanwhile, by integrating the spectral components from 390.6 to 392 nm, the corresponding normalized lasing intensity as a function of the gas pressure is also given. As can be seen, the maximum lasing intensity and spectral width are, respectively, achieved at the pressure of 20 and 60 mbars.

IV. DISCUSSIONS

To understand the above observations, we start our discussions with a simple case of N_2^+ fluorescence that is commonly generated in a femtosecond laser-induced plasma column or filamentation. It has been revealed by several groups that the radiation lifetime of the electronically excited state $B^2 \Sigma_u^+(\nu = 0)$ of N_2^+ measured by a streak camera is greatly reduced in comparison to its natural emission lifetime [27,28], which is typically ~ 60 ns [29]. Lei *et al.*, ascribed this to the ultrafast electron-ion collision occurring during the fluorescence emission after precluding the superradiant decay and nonradiative processes [27]. The accelerated depopulation rate of the excited electronic state $B^2 \Sigma_u^+(\nu = 0)$ results in a sharp fluorescence signal decay in time domain and a broadened spectral width on the basis of the natural width. So the total spectral width Γ of N_2^+ fluorescence can be expressed as

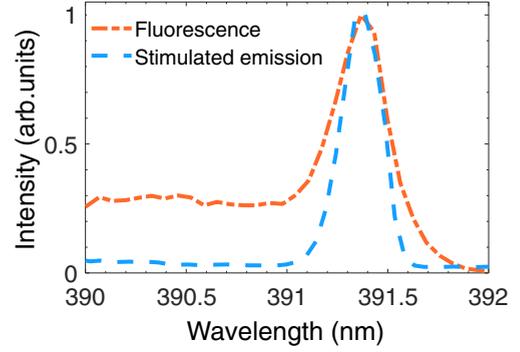


FIG. 4. Comparison of spectral widths of the measured 391.4-nm fluorescence and stimulated coherent emission at the pressure of 10 mbars.

$\Gamma = \Gamma_n + \Gamma_c$, where Γ_n is the natural width of the $B^2 \Sigma_u^+(\nu = 0)$ state, and Γ_c is the spectral broadening brought by collisions. In Fig. 4, we show the measured typical spectra of 391.4-nm stimulated emission (blue dotted curve) and noncoherent fluorescence (orange dashed-dot curve) of N_2^+ at the pressure of 10 mbars. It can be seen that the fluorescence spectral width is obviously larger than that of N_2^+ lasing, and the spectral widths of these two curves exceed its natural spectral width (not shown) of $B^2 \Sigma_u^+(\nu = 0)$ state governed by its natural lifetime. It is noteworthy that the spectral broadening of fluorescence stems from the energy-level shift caused by collisions.

The ultrafast electron-ion collisions under plasma condition could greatly accelerate the population decay of the upper level $B^2 \Sigma_u^+(\nu = 0)$, so how it critically influences the spectral and temporal widths of N_2^+ lasing relating to both the upper and the low levels of $B^2 \Sigma_u^+(\nu = 0)$ and $X^2 \Sigma_g^+(\nu = 0)$? We address the problem from the perspective of spectral broadening of N_2^+ lasing which reflects the time duration of gain since the lasing spectral width is inversely proportional to its time duration for the Fourier transform limit case. To analyze this, it is necessary to understand the gain mechanism underlying the N_2^+ lasing. The existing evidence manifests that the gain of N_2^+ lasing originates from both the population inversion Δn_{BX} between the states of $B^2 \Sigma_u^+(\nu = 0)$ and $X^2 \Sigma_g^+(\nu = 0)$ [17,23,24] and quantum coherence C_{XB} [9,19–21], both of which can be disturbed by collisions. Recently we show that for a large gain case, multichannel amplification routes including seed amplification, superradiance as well as free induction decay could interplay in this complicated process [13]. At the current position, we are only concerned with the temporal width T_ω of N_2^+ lasing under our experimental parameters to acquire a qualitative explanation for the aforementioned results. A crucial clue to address the spectral broadening of N_2^+ lasing is comparing its gain lifetime τ with the collisional time-span t_c . It has been verified that the temporal width T_ω of N_2^+ lasing within the range of a few to 10 ps depending on the gas pressure, nearly is inversely proportional to the gas pressure p , i.e., $T_\omega \propto p^{-1.2}$ [4]. Meanwhile, the collisional rate γ of electrons and ions can be evaluated by the equation $\gamma = \rho_e v_e \sigma$ [27], where ρ_e , v_e , and σ are, respectively, the electron density, electron velocity, and impact cross section. It has been estimated that at the pressure of about 8 mbars,

the collisional rate γ is about 10^{10} s^{-1} [27]. Based on this, we assume the collisional time-span t_c is about 100 ps for the pressure of 10 mbars and t_c is inversely proportional to the gas pressure.

The experimental results presented in Fig. 1 can be understood by the following. The gradual decrease in the N_2^+ lasing spectral width with the increasing pump-probe delay at the pressure of 40 mbars can be attributed to the continuous increasing of time-duration T_ω , according to the experimental results in the reference of Ref. [30] which demonstrated that the time duration T_ω of N_2^+ lasing increases with the pump-probe delay [30]. Actually, this is understandable theoretically with the aid of the superradiant picture that shows the radiation duration T_ω is inversely proportional to the population inversion density ρ [31], which declines with the increasing pump-probe delay due to the spontaneous emission and the collisional process. It deserves mentioning that only the electron-ion collisional process can match the gain lifetime τ of N_2^+ lasing, rather than the nonradiative collisions between ions and neutral molecules which takes place typically on nanosecond scale. Herein, we give a rough estimation of the electron-ion collisional rate. According to the previous measurement [23,32], the laser peak intensity is estimated to $2.9 \times 10^{14} \text{ W/cm}^2$, which causes nearly $\sim 10\%$ ionization probability and, hence, the electronic density at the pressure of 40 mbars reaches $\sim 5 \times 10^{16} \text{ /cm}^3$. Besides, the kinetic energy of the free electrons is 2 eV, and the cross-section σ is taken as the order of 10^{-14} cm^2 [27]. Substituting these parameters into the equation $\gamma = \rho_e v_e \sigma$, the resulting collisional time between the free electrons and the ions at the pressure is about 20 ps. This ultrafast collisions at the pressure could lead to a striking decrease of population inversion of the current ionic system. However, for the case of 10 mbars, the constant spectral width of N_2^+ lasing can be accounted for the invariable collisional factor within the whole pump-probe delays. Noted that at the pressure of 10 mbars, the gain lifetime τ is nearly twice that of it for the pressure of 40 mbars, which signifies the influence of the electron-ion collision can be milder in the case of 10 mbars. Therefore, as can be seen from the results in Fig. 1(a), it can be deduced that although the gain factor may be tinily reduced with the increasing pump-probe delays, the time-duration T_ω of N_2^+ lasing was barely changed due to little impact of electron-ion collisions at this pressure, resulting in a constant spectral width. This also predicts that the spectral width becomes broader as the pressure increases due to the shortened gain lifetime τ brought by the collisional processes as observed in Fig. 3.

For the experimental results shown in Fig. 2, it can be explained from the perspective of changing the probe intensity. Herein, changing the polarization angle θ is equivalent to varying the probe intensity along the pump polarization direction in terms of inducing laser gain. The prerequisite is that the molecular ions should possess a certain degree of molecular alignment at the delay avoiding the revival of molecular rotational wave packet, which can be attributed to permanent alignment of the molecules that caused by preferential ionization and anisotropic electronic coupling within the pump interaction. The permanent alignment effect in N_2^+ lasing is manifested by the dependence of the intensity and polarization direction of N_2^+ lasing on the polarization angle θ at

a nonalignment delay [2,9]. In Fig. 2, the main feature is that the spectral width narrows with the increasing polarization angle θ . Recall that the amplification mechanism responsible for N_2^+ lasing is superradiance in essence [13,15,19,20] in which the weak probe pulse acts as an external trigger. According to our recent experimental and theoretical results [30], it is easily known that both the retarded delay time T_D and time-duration T_ω increase with the decreasing probe intensity [13], thus, it explains the experimental observations in Fig. 2 since the probe intensity decreases with the growing polarization angle θ . Physically, this is because when a weaker probe pulse is injected in, the longer delay time T_D for establishing the macroscopic quantum coherence among the emitters is required and longer time-duration T_ω is needed to exhaust the gain of N_2^+ system due to the diminished quantum coherence in the presence of collisions. In a word, for the cases of 20, 40, and 60 mbars, the time-duration T_ω of N_2^+ lasing increases with the polarization angle θ , which causes that the measured spectral width decreases with the growing value of θ . For the pressure of 10 mbars, the spectral width hardly varies with the parameter θ due to the negligible collisional impact in terms of altering the time-duration T_ω of N_2^+ lasing as implied by the above analysis. Furthermore, it is expected that the molecular alignment caused by the revivals of a rotational wave packet could result in a more obvious dependence of laser gain on the polarization angle θ in comparison to that induced by permanent alignment. The quantitative relation of molecular alignment to polarization dependence is studied in progress.

Finally, we would like to point out that another element that could bring some uncertainty for the absolute spectral width of N_2^+ lasing is the instrumental broadening since the measured spectral profile is generally the convolution of the actual spectral profile and the instrument response function whereas the random process during the measurement is ignored, i.e., $f(\nu) = f_T(\nu) \otimes f_S(\nu)$ [33], where $f(\nu)$, $f_T(\nu)$, and $f_S(\nu)$ are, respectively, the measured spectral, the true spectral, and the instrumental profiles. The spectral resolution of our current installation is about 0.48 nm. Due to the limited resolution, the measured spectral width of N_2^+ lasing may be influenced by the instrumental aspects. The spectral resolution can be improved by the use of a more advanced spectrometer [6,34]. Fortunately, it can be inferred from the above equation that for an invariable instrumental response, the positive relation between the observed and the true spectral widths (i.e., the wider of the true spectral width, the wider of the measured spectral width) validates the current qualitative analysis on the spectral width of N_2^+ . Therefore, the instrumental spectral broadening could not change the unraveled physics of the present paper.

V. CONCLUSIONS

In conclusion, we investigated the spectral width variation of N_2^+ lasing created in a typical weakly ionized plasma by monitoring the experimental parameters including the pump-probe delay, polarization angle θ , and gas pressure. It was found that at a relatively high pressure of 40 mbars, the spectral width of N_2^+ lasing obviously shortens as the pump-probe delay or polarization angle θ increases, which is distinctly

different from that at a low pressure of 10 mbars. From the perspective of considering the time duration of N_2^+ lasing, our analysis shows that the electron-ion collisional process play a primary role in determining the length of gain lifetime of N_2^+ lasing. It is anticipated that in the future the influence of ultrafast collisions on ionic coherent radiations can be separately investigated by varying the concentration ratio of a gas mixture, similar to the studies of ionic fluorescence [27] and harmonic-seeded ionic lasers [35]. Our findings benefit the understandings of the amplification mechanism of N_2^+ lasing in the facet of considering the effect of ultrafast electron-ion collisions, and they also shed light on the spectral manipulation of coherent emissions produced in plasma surrounding during strong laser-matter interaction by showing

difference with the well-known spectral control caused by molecular alignment.

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