Generation of Raman lasers from nitrogen molecular ions driven by ultraintense laser fields

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Abstract

Atmospheric lasing has aroused much interest in the past few years. The ‘air–laser’ opens promising potential for remote chemical sensing of trace gases with high sensitivity and specificity. At present, several approaches have been successfully implemented for generating highly coherent laser beams in atmospheric condition, including both amplified spontaneous emission, and narrow-bandwidth stimulated emission in the forward direction in the presence of self-generated or externally injected seed pulses. Here, we report on generation of multiple-wavelength Raman lasers from nitrogen molecular ions (N₂⁺), driven by intense mid-infrared laser fields. Intuitively, the approach appears problematic for the small nonlinear susceptibility of N₂⁺ ions, whereas the efficiency of Raman laser can be significantly promoted in near-resonant condition. More surprisingly, a Raman laser consisting of a supercontinuum spanning from ~310 to ~392 nm has been observed resulting from a series near-resonant nonlinear processes including four-wave mixing, stimulated Raman scattering and cross phase modulation. To date, extreme nonlinear optics in molecular ions remains largely unexplored, which provides an alternative means for air–laser-based remote sensing applications.

1. Introduction

The advent of ultrashort and ultraintense laser pulses has revolutionized the interaction of light with matter, giving rise to non-perturbative tunnel ionization which is the pillar stone of strong field laser physics [1]. With the tunnel ionization as the initiative process, highly nonlinear processes such as high-order harmonic generation [2], above threshold ionization [3], and non-sequential double ionization [4] have been observed, which further provide the means to access the dynamics in atomic and molecular systems on attosecond time scale. Recently, lasing actions induced by tunnel ionization of nitrogen molecules have been observed which come as a major surprise to those who have been investigating strong field physics over the past three decades [5–23]. These observations were made with either a pump laser at 800 nm wavelength [6, 8–21] or that at longer wavelengths in the range between 1 and 4 μm [5, 7, 23].

Interestingly, further pump–probe investigations on the generation of N₂⁺ lasers at 391.4 nm, which corresponds to the transition between N₂⁺(B²Σ⁺u⁺, ν’ = 0) and N₂⁺(X²Σ⁺g⁺, ν = 0) states, clearly show that the gain of the external seed pulses whose spectra overlap the 391.4 nm transition line can only be observed for the pump laser at 800 nm wavelength but not at the other pump wavelengths in the infrared range [17]. The experimental results lead to two consequences as follows. First, N₂⁺ ions produced in the 800 nm fields are preferentially populated on the excited N₂⁺(B²Σ⁺u⁺, ν’ = 0) state but not the ground N₂⁺(X²Σ⁺g⁺, ν = 0) state, resulting in a population inversion which is difficult to understand in the framework of tunnel ionization [1]. At
present, several mechanisms have been proposed for revealing the origin of the population inversion, including the depletion of ground state via one-photon absorption [17, 18, 20], field-induced multiple recollisions [15, 21], transient inversion between rotational wavepackets of excited and ground states [22], different rotational distributions in two states [23], and so on. Nevertheless, a final conclusion is yet to be drawn. Second, there should be no population inversion in $+^{1}N_2$ ions produced in laser fields at infrared wavelengths ranging from 1 to 2 $\mu$m as evidenced by the failure of observing any gain in the external seed pulses injected immediately after the pump pulses to avoid temporal overlap between the two [17]. The origin of the laser-like emissions observed only with the self-generated seed pulses (i.e., the harmonics of the pump laser) at pump wavelengths longer than 1 $\mu$m is another puzzle yet to be unlocked.

To understand how the $+^{1}N_2$ laser-like emissions are generated with the pump pulses of longer wavelengths, we perform a systematic investigation on the generation of $+^{1}N_2$ coherent emissions at various pump wavelengths in the range of 1.2 and 2 $\mu$m, which is the wavelength range of our OPA source. Surprisingly, our observations indicate that some conclusions drawn from the previous experiments are not always true. Among the deviations, the most significant two features reported here are (1) generation of the $+^{1}N_2$ laser-like emission can be achieved even when the spectra of seed pulses do not overlap the observed $+^{1}N_2$ transition lines, and (2) the narrow-bandwidth $+^{1}N_2$ emission can be significantly broadened to form a supercontinuum-like spectrum with a bandwidth of $\sim$80 nm at low gas pressures. The finding shows that Raman lasers can be efficiently generated in $+^{1}N_2$ ions in the near-resonant condition using intense mid-infrared pump lasers, which has important implication for realizing air lasing.

2. Experimental setup

The schematic diagram of the experimental setup is shown in figure 1. The experiments were carried out using an optical parametric amplifier (OPA, HE-TOPAS, Light Conversion Ltd), which was pumped by a commercial Ti:sapphire laser system (Legend Elite-Duo, Coherent, Inc.). The OPA enables to generate wavelength-tunable femtosecond laser pulses in the range from 1.2 to 2.4 $\mu$m at a repetition rate of 1 kHz. A dichroic mirror (i.e., Filter 1) was used to remove the weak visible light around 400 nm from OPA. The pump laser pulses were then focused into the gas chamber filled with nitrogen gas using an $f = 10$ cm lens, and were further collimated using another lens with the same focal length. The signal beam exiting from the gas chamber together with the pump laser were reflected by an uncoated glass plate, and the residual pump laser was eliminated with a piece of blue glass (i.e., Filter 2). At last, the generated third harmonic (TH) and fifth harmonic (FH) beams were focused into a grating spectrometer (Shamrock 303i, Andor) by a lens with a focal length of 15 cm. A neutral density filter was used to avoid saturation of the detector. The spectra of the fundamental wave (FW) after passing through the gas chamber were measured by a fiber spectrometer (NIRQuest512, Ocean Optics, Inc.) with an integration sphere.

3. Experimental results

3.1. Generation of coherent emissions in $+^{1}N_2$ with mid-infrared pump lasers

Figure 2(a) shows the dependence of FH spectra on the gas pressure of nitrogen molecules obtained with pump pulses at a wavelength of 1580 nm. The pump power is approximately 1.0 W, and the beam diameter ($1/e^2$ width) is 7.6 mm. The pulse duration is measured to be $\sim$60 fs. At the pump wavelength, neither the third nor...
the FH of the pump laser could cover the transition lines at either 358.2 nm or 391.4 nm wavelengths. Surprisingly, two strong coherent emissions appear near the two transition lines at 358.2 nm and 391.4 nm wavelengths, which correspond to $S_{\epsilon} \rightarrow S_{\epsilon}^{\pm}$.

$N^+_2 (B^2\Sigma_u^+, \nu' = 1) \rightarrow N^+_2 (X^2\Sigma_g^+, \nu = 0)$ and $N^+_2 (B^2\Sigma_u^+, \nu' = 0) \rightarrow N^+_2 (X^2\Sigma_g^+, \nu = 0)$, respectively. Meanwhile, a very weak emission at ~331 nm also appears on the FH spectrum, which corresponds to the transition $N^+_2 (B^2\Sigma_u^+, \nu' = 2) \rightarrow N^+_2 (X^2\Sigma_g^+, \nu = 0)$. The laser-like emissions show a small divergence half-angle of ~15 mrad. This divergence angle can be further reduced in the looser focusing condition if the pump laser could provide higher output power. Furthermore, the spatial and temporal coherence of the laser-like emissions is confirmed by interferometric measurements, in which the interference fringes created in spatial and temporal domains are clearly observed.

In previous reports [5, 7–21], $N^+_2$ coherent emissions from different vibrational transitions between

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Figure 2. Dependences of the fifth harmonic spectra generated in (a) nitrogen and (b) argon gases with the 1580 nm pump laser on gas pressures (logarithmic color scale). (c) Fifth harmonic spectra obtained in nitrogen and argon at a gas pressure of 20 mbar. To facilitate a quantitative comparison, the harmonic spectrum of argon in figure 2(c) is multiplied by a factor of 10.

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6 See supplementary material for more details which is available online at stacks.iop.org/NJP/20/033035/mmedia.
ground and excited states show an extremely narrow spectral bandwidth. To our knowledge, this is the first observation of coherent supercontinuum generated in \( \text{N}_2^+ \) ions at low gas pressures. The gas pressure required for generating the supercontinuum decreases with the increase of the focal length of lens, implying that high laser intensity is necessary for generating the supercontinuum (see footnote 6). When the nitrogen molecular gas pressure increases to ~40 mbar, the supercontinuum disappears from the measured spectra, and the narrowbandwidth emissions at ~358 and ~391 nm show a rapid decay with the increase of gas pressure, as shown in figure 2(a). We believe that this is due to strong plasma defocusing at relatively high gas pressures, which will prevent the build up of high peak intensity near the focus.

Naturally, one would expect that the supercontinuum generation near the transition lines of \( \text{N}_2^+ \) ions is a characteristic related to its energy level structure. We have confirmed this by carrying out a comparative experiment in argon, as argon has an ionization potential and a nonlinear coefficient similar to that of nitrogen molecules [24]. In this case, all of the experimental conditions remained the same except that the gas in the chamber was changed from nitrogen molecules to argon atoms. The FH spectrum generated in argon as a function of the gas pressure is shown in figure 2(b), which shows a smooth spectrum with a reasonable bandwidth of 9–10 nm (FWHM) and centered at the wavelength of ~319 nm (i.e., close to one fifth of the pump wavelength). The bright narrow-bandwidth emissions as well as the supercontinua both disappear in argon, as shown in figure 2(b).

To facilitate a quantitative comparison, figure 2(c) shows the FH spectra measured in nitrogen and argon at a gas pressure optimized for the observed strong supercontinuum generation (i.e., 20 mbar). In comparison with argon, the harmonic signal generated in nitrogen shows two striking differences. First, the FH generated in nitrogen covers an extremely broad spectral range with strong \( \text{N}_2^+ \) laser-like emissions superimposed on the supercontinuum spectrum. Second, the FH produced in nitrogen shows pronounced red shift in its spectrum and enhancement of the signal intensity by approximately one order of magnitude in comparison with the FH generated in argon. These differences could be due to the different origins of harmonics, namely, the FH generated in argon is mainly from the neutral atoms whereas the FH generated in nitrogen molecules is mainly from the molecular ions.

An immediate thought after seeing the spectrum in figure 2(a) would be that such unusual FH spectra should be a result of the strong distortion in the spectrum of pump pulses and/or that in the spectrum of the TH wave caused by nonlinear propagation of the pump laser pulses. Therefore, we measured the spectra of the pump pulses and its TH in both nitrogen and argon gases at different gas pressures. As shown in figures 3(a)–(d), both the spectra in nitrogen and argon are very similar without any noticeable differences. From figures 3(a) and (c), one can see that below 30 mbar, the spectra of the pump pulses are undergoing a continuous blue shifting as well as a spectral broadening with the increase of the gas pressure. The blue shift can be attributed to the plasma generation as have been intensively discussed in previous investigations [25]. Above 30 mbar, the spectral broadening becomes more pronounced than the blue shift. For the TH signals, the intensities increase with the increasing gas pressure, and the spectra broaden as well, as shown in figures 3(b) and (d). Nevertheless, both the pump laser pulses at the FW and the generated TH signal show limited increases in the spectral bandwidths. Thus, we exclude the possibility of generating the supercontinuum via FH generation driven by the mid-infrared pump pulses or four-wave mixing utilizing the fundamental pump pulse and its TH.

### 3.2. Dependence of supercontinuum emission in \( \text{N}_2^+ \) on laser parameters

To gain insight into the physical mechanism behind the result in figure 2(a), we further investigate the dependences of the FH signals generated in nitrogen molecules on the intensity, polarization and wavelength of the pump laser. Figure 4(a) shows the supercontinuum spectra measured at different pump laser intensities for a fixed gas pressure of 16 mbar. The supercontinuum already began to appear at a relatively low pump power of 200 mW, corresponding to a peak intensity of \( \sim 1 \times 10^{15} \) W cm\(^{-2}\) as evaluated by assuming linear propagation of the Gaussian pump laser in the gas chamber. The assumption of linear propagation can be justified with the low gas pressure. When the pump laser intensity increased, the supercontinuum became stronger and its spectrum became broader. Then, we changed polarization of the pump laser at an average power of \( \sim 1 \) W from linear to circular polarization using an achromatic quarter-wave plate at the gas pressure of 16 mbar. Here, the angle of 0° corresponds to linear polarization, and the angles of ±45° correspond to circular polarization. Since the reflection from the uncoated glass plate is sensitive to the polarization of the incident light, we replaced the uncoated glass plate with a reflective mirror which is insensitive to the laser polarization. As shown by figure 4(b), the supercontinuum-like emission generated with the linearly polarized pump lasers is the strongest in its intensity and the broadest in its spectrum, which is different from the result obtained with 800 nm pump laser [15]. In contrast, changing the polarization of pump pulses from linear to circular polarization leads to a significant drop of the signal intensities by 3–4 orders of magnitude. By comparing figures 4(a) and (b), we can confidently confirm that the disappearance of both the supercontinuum and \( \text{N}_2^+ \) emissions in the circularly polarized driver laser fields cannot be completely attributed to the lowering of field strength with the circular...
Figure 3. Dependences of the spectra of (a) the fundamental wave at 1580 nm wavelength after passing through gas chamber and (b) the generated third harmonic on gas pressures of nitrogen gas. For comparison, the corresponding results obtained from argon gas with the same laser conditions are present in (c) and (d), respectively. All figures are present with a logarithmic color scale.

Figure 4. Dependence of the fifth harmonic spectra generated in nitrogen gas at a gas pressure of 16 mbar on (a) intensity and (b) polarization of the 1580 nm pump laser, and the pressure-dependent fifth harmonic spectra generated by the pump laser at (c) 1780 nm and (d) 1400 nm wavelength. All figures are present with a logarithmic color scale.
polarization. In addition, the fact that the FH, the laser-like emissions and the supercontinuum all depend on the polarization of the pump laser in a similar way provides clear evidence that their generation mechanisms are not independent but closely related to each other.

Another important feature of the supercontinuum generation is its strong dependence on the pump wavelength. To clarify this, we examine the supercontinuum generation at various mid-infrared pump wavelengths. Here, we present the measurement results at two typical pump wavelengths of 1780 nm and 1400 nm in figures 4(c) and (d), respectively. The pump powers at 1780 nm and 1400 nm were approximately 0.83 W and 1.0 W, respectively, which are the maximum output powers of our OPA system. The beam diameters were measured to be 9.3 mm and 6.2 mm for the 1780 nm and 1400 nm pump laser, respectively, and their pulse durations are close to that of 1580 nm (i.e., ~60 fs). It was observed that at the pump wavelength of 1780 nm, the coherent emissions at ~391 nm are both efficiently generated as evidenced by figure 4(c), with a supercontinuum filling up the gap between the two emission lines. Although strong coherent emission at ~428 nm corresponding to the transition $N^2_2 (B^2Σ_u^+, ν = 0) → N^2_2 (X^2Σ_g^−, ν = 1)$ is also observed, no supercontinuum is observed between the laser lines at ~391 and ~428 nm. Meanwhile, some weak emissions corresponding to the transitions between the high vibrational energy levels of $N^2_2 (B^2Σ_u^+) and$ that of $N^2_2 (X^2Σ_g^−)$ states appear in the spectral range of 410–420 nm. In contrast, switching the pump wavelength from 1780 to 1400 nm leads to elimination of the coherent emissions at ~391 and ~358 nm as well as the supercontinuum in between, as shown in figure 4(d). In such a case, only a strong emission at ~428 nm appears on the TH spectrum. The results in figures 2(a), 4(c) and (d) indicate the important role of the pump wavelength in the generation of the supercontinuum and $N^2_2$ coherent emissions.

4. Discussion

The key message from the results in figures 2 and 4 is that the $N^2_2$ coherent emissions at ~391 and ~358 nm wavelengths can only be achieved when the spectra of FH overlap at least one of the transition lines between $N^2_2 (B^2Σ_u^+) and N^2_2 (X^2Σ_g^−)$ states. This feature suggests that the FH generation can be enhanced as a result of some resonant nonlinear processes. Although the FH can be generated through either a direct fifth-order nonlinear process (i.e., $5ω = ω + ω + ω + ω + ω$) or a cascaded third-order process (i.e., $5ω = 3ω + ω + ω$), the latter should play a dominant role for our case (see footnote 6). The resonant four-wave mixing process is schematically depicted in figure 5(a). We would like to stress that due to the high peak intensity of pump pulses, the resonant process can be very efficient. As shown in figure 2(c), the resonantly generated FH spectrum is about one order of magnitude stronger than that in argon, and the resonant process forces the central wavelength of the FH to shift to near 331 nm.

Subsequently, the generated $5ω$ photons at ~331 nm can initiate the stimulated Raman scattering (SRS) in the excited $N^2_2$ ions, which leads to the generation of photons at ~358 nm, as depicted in figure 5(b). Once the coherent emission at ~358 nm is generated, it triggers further SRS process to produce the photons at ~391 nm, as depicted in figure 5(c). In general, Raman scattering as illustrated in figures 5(b) and (c) requires an initial population in $N^2_2 (B^2Σ_u^+)$ state [26]. In our experimental conditions (i.e., tight focusing geometry, high power and low pressure), a decent amount of ions can be populated to $B^2Σ_u^+$ state under such a laser condition [17]. The population on $B^2Σ_u^+$ state can be further promoted with the resonant single-photon absorption of FH photons. One concern is that the FH generated in $N^2_2$ ions might be too weak to initiate SRS processes. To understand this, it should be noticed that the resonance of the Stokes photons generated by the Raman process in figures 5(b) and (c) with the electronic states of $N^2_2$ ions will efficiently promote the gain and meanwhile reduce the threshold of SRS [27]. Moreover, the Raman process can be more efficient for the vibrational ground state because of the larger population, smaller damping rate, and larger dipole moment in the vibrational ground state. In such a way, the ~331 nm photons produced in a traveling pump regime are efficiently converted to ~358 nm and then ~391 nm photons. This is why the $N^2_2$ emission line at ~391 nm is the strongest even though the harmonic spectrum cannot cover the transition line. Some more rigorous analysis can be found in the supplementary material (see footnote 6).

The last question is how the supercontinua between the stimulated Raman lasing lines are generated although they are not resonant with any transitions between $N^2_2 (X^2Σ_g^−)$ and $N^2_2 (B^2Σ_u^+)$ states. It is noteworthy that rotational transitions between the two electronic states cannot cover such a broad spectral range [9, 14, 23]. On the other hand, as we have discussed above, high peak intensities can be reached in our experimental condition. Therefore, cross phase modulation can occur immediately after the generation of the stimulated Raman lasers in the intense pump laser field, as illustrated in figure 5(d). The cross-correlation measurement also shows that the strong $N^2_2$ emission driven by the mid-infrared pump laser mainly occurs within the pulse duration of the mid-infrared pump laser, which is totally different from that with the 800 nm pump laser [8, 15]. The temporal overlap of two pulses enables the cross phase modulation between these narrow-bandwidth $N^2_2$ emissions and the intense pump laser. In particular, the Raman lines near 358 and 391 nm wavelengths are in
resonances with the transitions between $\text{S}^{\text{++}}(\text{X}^2\Sigma_g^+)$ and $\text{S}^{\text{++}}(\text{B}^2\Sigma_g^+)$ states. In such a case, the nonlinear Kerr coefficient will be greatly enhanced, leading to the extraordinary supercontinuum generation which is rarely observed with atomic or neutral molecular gases under the similar experiment conditions. In addition, because the cross phase modulation mainly occurs in the falling edge of the pump pulse, the spectral blue shift is much more pronounced than red shift \[28\], as evidenced by our experimental observations in figure 2(a). The physics involved in the processes above is discussed more thoroughly in the supplementary material (see footnote 6).

When the pump wavelength is switched to 1780 nm, a nonlinear process similar to that in figures 5(a)–(d) would dominate the generation for both the lines at $\sim 358$ and $\sim 391$ nm wavelengths as well as the supercontinuum in between. For the pump laser at 1400 nm, although its TH spectrum can cover the transition line at $\sim 428$ nm, a low population in $\text{S}^\text{=} (\text{v} = 1)$ state makes it difficult to excite both SRS and cross phase modulation (see footnote 6). As a result, the supercontinuum emission is hardly observed at the pump wavelength, which is in good agreement with the experimental observation in figure 4(d). In addition, it should be emphasized that the supercontinuum generation results from resonant nonlinear processes in molecular ions, so that the pump laser must be sufficiently strong to generate a large amount of ions in both excited and ground states. Otherwise, the nonlinear optical signals generated in neutral molecules will smear the signals generated in the molecular ions. Thus, supercontinuum emission in $\text{N}_2^+$ ions shows a strong dependence on gas pressure, focal geometry, and the pump intensity, as illustrated in figures 2(a), 3 and 4(a) (see footnote 6). Meanwhile, all resonant three-order nonlinear processes involved in the picture will not occur in a circularly polarized laser field \[29\]. This is why both the $\text{N}_2^+$ laser lines as well as the supercontinuum disappear for the circular polarization. Therefore, the picture presented above provides a qualitative but logical explanation on all the experimental observations. Quantitative analyses on these processes require calculation of nonlinear susceptibilities of multiple vibrational and rotational states of $\text{N}_2^+$ ions and numerical simulations on the nonlinear propagation in $\text{N}_2^+$ ions, which will be an interesting challenge for the theorists of strong field molecular physics.

It is of interest to determine the conversion efficiency of the laser-like emissions. The average power of the 391 nm emission generated at the gas pressure of 20 mbar was measured to be $\sim$ 3 nW. Therefore, the conversion efficiency from the 1580 nm pump pulse to the 391 nm emission is determined to be $\sim 3 \times 10^{-9}$, which appears low. However, we should notice that the 391 nm emission originates from the SRS process in which the driver
field is the FH of the 1580 nm pump laser. Based on this picture, the conversion efficiency of 391 nm laser-like emission from the FH must be several orders of magnitude higher than $\sim 3 \times 10^{-4}$. Due to the continuous conversion from the FH to the 391 nm emission along the plasma channel, it is difficult to precisely determine the conversion efficiency of the FH from the 1580 nm pump laser field. Nevertheless, from the spectrum in figure 2(c), a large portion of the FH has been converted to the laser-like emission at 391 nm as well as the supercontinuum, indicating a very efficient wavelength conversion process.

5. Conclusions

To conclude, we have investigated near-resonant extreme nonlinear optics in nitrogen molecular ions at several wavelengths between 1.2 and 2 $\mu$m. Due to the existence of abundant vibrational energy levels, a series of third-order nonlinear optical processes including three-photon excitation, SRS, and cross phase modulation can occur almost simultaneously at multiple resonant wavelengths. We notice that nitrogen molecular ions are a unique quantum system for demonstrating such extreme nonlinear effects. Since these molecular ions have a large ionization potential as high as $\sim 1$ eV, they can survive against photoionization at visible and infrared wavelengths even when the pump laser intensity reaches $8.5 \times 10^{14}$ W cm$^{-2}$ (assuming an ionization probability of $\sim 1$%). In the meantime, the nitrogen molecular ions can also allow the transitions between various vibrational levels of $\text{N}_2^+$ ($B^2\Sigma_u^+$) state and $\text{N}_2^+$ ($X^2\Sigma_g^+$) state to occur at near ultraviolet wavelengths in the near-resonant condition. The characteristics are of vital importance for generating atmospheric Raman lasers.

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