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非相干电离注入导致自由感应衰变信号的擦除效应

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摘　要:由强场中红外飞秒脉冲泵浦的氮离子能够以自由感应衰变(FID)的形式发出相干的前向辐射,该FID辐射可以被随后的800 nm飞秒脉冲有效抑制。这种擦除效应是对微弱FID辐射进行时间表征的独特工具,特别是在FID辐射与泵浦脉冲的谐波在频域重叠的情况下。基于密度矩阵与麦克斯韦方程的数值模拟结果,认为擦除效应主要是由于800 nm脉冲直接引发氮离子B态和A态相干性的变化,该变化进一步与800 nm脉冲耦合作用,引起了B态和X态之间相干性的减小,从而导致对应的391.4辐射的减弱。

关键词:空气激光;超快光谱技术;自由感应衰变;飞秒脉冲
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0 Introduction

Nitrogen molecules illuminated by an intense femtosecond pulse in the near-infrared or mid-infrared regime give rise to coherent forward emission at a series of wavelengths, including 391.4, 427.8, 357.8 nm, etc [1]. Because optical amplification of an external seeding pulse may appear in such nitrogen gas plasma pumped by 800 nm pulses, this effect together with other optical amplification effects inside air plasma has been coined as "air lasing"[2-4]. These cavity-free air lasing effects have attracted many interests in recent 10 years since they hold the unique potential to generate a remote virtual lasing source in ambient air, which is expected to bring revolutionary improvement in optical remote sensing[2,5-6]. Recently, it has been demonstrated that lasing emission of neutral or ionic nitrogen molecules can serve as the light source for detection of gas with ppm sensitivity[6]. Concerning the underlying mechanism for the coherent emission of neutral N_2, it is now well accepted that the 337.4 nm emission is Amplified Spontaneous Emission (ASE) and the population inversion between the third electronic excited state $C^3\Pi_u^+$ and second excited state $B^3\Pi_g^+$ has been established[7]. In contrast, the mechanism for lasing of N_2^+ is much more complicated and it is still under intense debate[8-14]. This debate stems partially from the mysterious fact that the coherent 391.4 and 427.8 nm emission can always be observed regardless the pump laser wavelength is 400 nm, 800 nm, 1 100 to 1 900 nm, or even 3 900 nm[15-19]. Very recently, it became clear that the nature of the 391.4 and 427.8 nm emission is distinct when pump lasers

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with different wavelengths were used as the driving pulse[10,20-21].

When tunable MIR femtosecond pulses were employed as the pump laser, it has been observed that coherent forward emission at a series of wavelengths including 391.4, 427.8 nm superposed on the third or fifth harmonics of the pump laser[1,21-22]. It was now recognized that the nature and mechanism of the 391.4 and 427.8 nm emission in case of MIR pumping is different from that of 800 nm pumping[10,21]. A significant difference between the 391.4 nm emission pumped by MIR pulses and the 800 nm pulses lies in the fact that in the former case no optical amplification occurs when external seeding pulses were injected into the plasma[10]. At the same time, due to the limited pulse energy of the MIR pump pulses used in different laboratories (typically 0.1～0.5 mJ), the forward coherent 391.4 or 427.8 nm emission is much weaker than their counterpart in case of 800 nm pump pulse with typical pulse energy of 1～10 mJ. Recently, the temporal profile of coherent 391.4 nm emission pumped by intense MIR pulses was characterized with Sum-Frequency Generation (SFG) technique, in case that the 391.4 nm signal was well separated from the 3rd or 5th harmonic when some particular pump laser wavelengths were chosen[21]. Based on the spectral and temporal features, the 391.4 nm emission obtained with MIR pumping has been identified as Free Induction Decay (FID) where the resonant excitation of the B-X coherence by the tunable MIR laser pulse plays an important role [21]. However, in most of the case with the mid-infrared pump pulses, the 391.4 nm emission was superposed on the harmonics in the spectral domain[1,10,22]. The spectral overlap of the 391.4 nm radiation with the harmonics, together with its relatively weak intensity, hinders its measurement with the nonlinear SFG technique and other method for temporal characterization is highly desired.

In this work, we injected a delayed 800 nm pulse into the plasma after the main mid-infrared pump pulse and observed an erasing effect of the free-induction decay emission. It was found that the erasing effect lasts for a couple of picoseconds for the pump wavelengths of 1 250 and 1 550 nm. In particular, for the pump pulse at 1 550 nm, a significant enhancement of the emission around temporal overlapping of the pump and control pulses was observed, while for the 1 250 nm pump pulse such enhancement was not found. Furthermore, this erasing effect was studied for different gas pressures and control pulse energies. We attribute this erasing effect to the incoherent ionization injection of 800 nm control pulse and the change of ρ_{BX} coherence in the system caused by the 800 nm pulse. This erasing effect provides a simple method for temporal characterization of the weak FID emission in case it overlaps with the harmonics in the spectral domain.

1 Experimental setup

In the experiment, a femtosecond laser system (Coherent Legend DUO) delivers 40 fs pulses with pulse energy of 12 mJ at a repetition rate of 1 kHz. A dielectric beam splitter separates the pulses into two beams of almost equal energy. One of the beams with 5 mJ pulse energy was employed to pump an Optical Parametric Amplification (OPA) system, which provides wavelength-tunable femtosecond pulses ranging from 1 100 nm to 2 600 nm. The other 800 nm beam was attenuated properly before it was combined with the MIR pump by a dichroic mirror. The experimental setup is schematically presented in Fig. 1. Both the MIR pump laser and the 800 nm control pulse were linearly polarized in the same direction, since it is known that the emission was optimized for a linearly polarized MIR pump pulse[22]. An optical delay line was mounted into the 800 nm control pulse in order to vary the time delay τ_d between the MIR pump and 800 nm control pulse. The collinear MIR and 800 nm beams were focused into a gas chamber filled with nitrogen by a convex lens of $f=30$ cm or 50 cm (LA1256-C or LA1380-C, Thorlabs). In our experiments, the pulse energy of the MIR pump pulse was 400～600 μJ, while the energy of the 800 nm control pulse was 200～300 μJ. Both the MIR pump and the 800 nm control pulse were capable to produce a visible plasma in the gas chamber. At the exit of the gas chamber, the forward emission from the gas plasma was collected into a fiber by a lens of $f=10$ cm (LA4545, Thorlabs) for spectral analysis with a spectrometer (HR 4 000, Ocean Optics). Several shortpass filters (BG 40) were installed before the fiber for proper spectral filtering. In the experiment, the forward spectrum from the plasma in the UV range was recorded as a function of the time delay between the pump and control pulses.

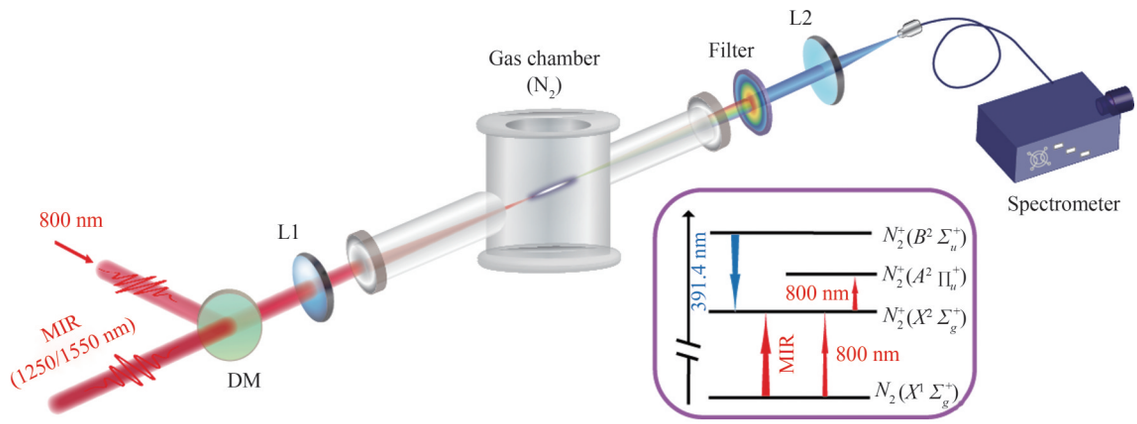
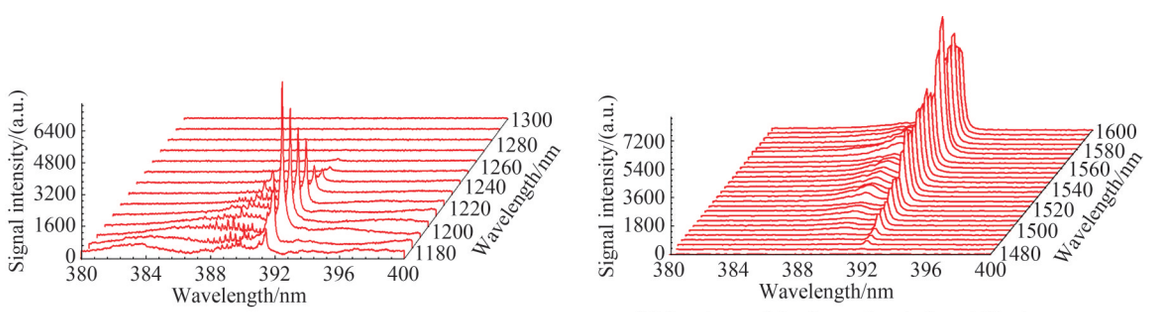


Fig.1 Experimental setup. Inset, energy-level diagram of neutral and ionic nitrogen molecule

2 Experimental results

2.1 Lasing emission at 391.4 nm from molecular nitrogen ion

In Fig. 2, the forward emission spectra from the nitrogen plasma were presented for tuned pump wavelength. For the pump laser wavelengths investigated, a narrow and sharp emission peak at 391.4 nm and a series of spectral peaks around 389 nm were observed, which correspond to the P and R branches of the transition of the nitrogen ions from the second excited state $B^2\Sigma_u^+(\nu'=0)$ to the ground state $X^2\Sigma_g^+(\nu=0)$. This emission has been observed in several previous reports and has been recently attributed to free-induction decay enabled by resonant excitation with the MIR pump pulses[1,10,21]. In case of pump wavelength λ_p = 1 250 nm, the sharp 391.4 nm peak was found to superpose on the relatively broad 3rd harmonic emission spectrum. While for 1 550 nm pump laser, the FID emission is clean in the spectral domain since both the 3rd and 5th harmonics are located away from the spectral peak.



(a) Spectrum of the forward emission at 30 mbar pressure and 460 μJ pump energy

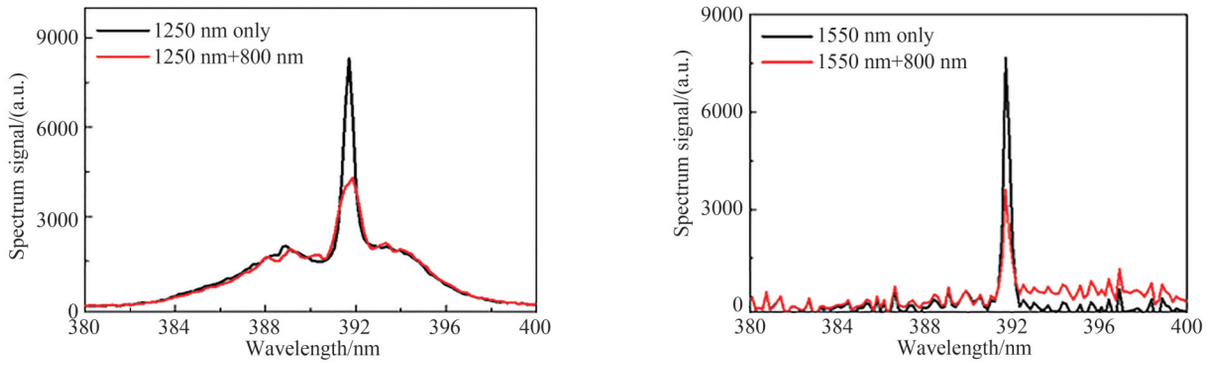
(b) Spectrum of the forward emission at 80 mbar pressure and 330 μJ pump energy

Fig.2 Spectrum of the forward emission as a function of the pump laser wavelength

2.2 The erasing effect by 800 nm control pulses

In Fig. 3, we present the emission spectra with the injection of a subsequent 800 nm control pulse for pump wavelength of 1 250 nm and 1 550 nm. The time delay between the MIR pump and the 800 nm control pulse were τ_p = 1 ps. We observed that the FID emission in both cases was substantially suppressed. Interestingly, we noticed that the relatively broad 3rd emission in Fig. 3(a) remains untouched. This indicate that the FID emission lasts for more than 1 ps while the duration of the 3rd harmonic is less than 1 ps since it is not changed.

The time-resolved results for pump laser wavelength of 1 250 nm and 1 550 nm are presented in Fig. 4 and Fig. 5. Here the positive delay corresponds to that the control pulse lags the MIR pump pulse. In Fig. 4 (a), it was observed that the 391.4 nm FID signal experiences a sudden decrease around the τ_d = 0 and recovers to its initial level after a delay of about 3 ps, regardless of the gas pressure. This indicates that for gas pressure ranging from 50 mbar to 200 mbar the FID signal presents a similar duration of 3 ps. We note that this feature is

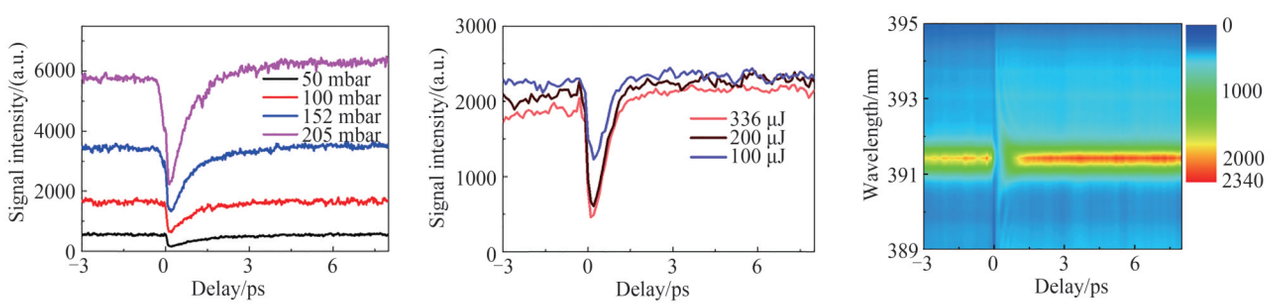


(a) Characteristics of emission at 391.4 nm for pump wavelength of 1250 nm

(b) Characteristics of emission at 391.4 nm for pump wavelength of 1550 nm

Fig.3 Suppression of the emission obtained at different pump wavelength

in sharp contrast with the superradiance of nitrogen ions where the emission duration is inversely proportional to the gas pressure[8]. We also varied the energy of the control pulse and the results are presented in Fig. 4(b). It was found that the FID signal was suppressed by about 75% for control pulse energy of 300 μJ and 200 μJ, while for 100 μJ pulse the FID signal is less suppressed. This is understandable since the higher energy control pulse results in much ionization, which will be discussed later. In Fig.4 (c), we presented the emission spectrum as a function of the time delay. It is observed that the entire free induction decay emission was suppressed for a time interval of about 3 ps.

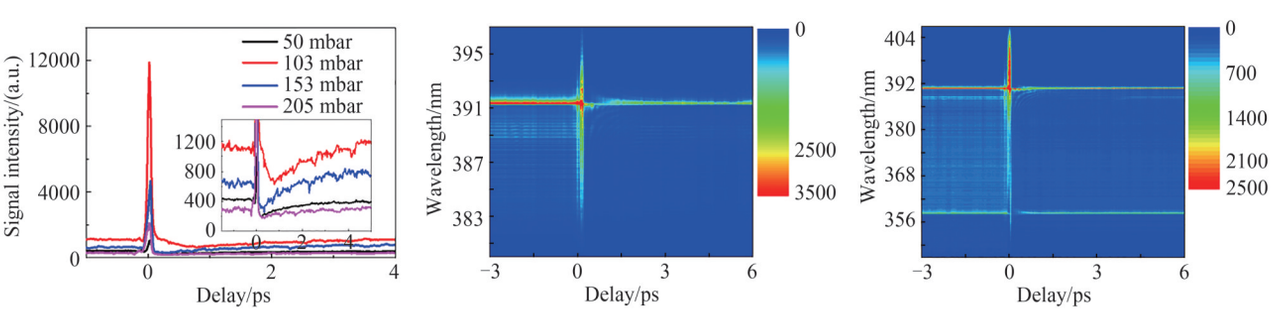


(a) Spectrum intensity as a function of the time delay for four different gas pressures

(b) Spectrum intensity as a function of the different contorl pulse energies

(c) The time-resolved measurement of the entire forward radiation

Fig.4 Experimental results of 1 250 nm pump wavelength



(a) Spectrum intensity as a function of the time delay for four different gas pressures, the inset is the zoom to show the details

(b) Time-resolved measurement of the forward radiation for 1550 nm pump (360 μJ) with 800 nm control pulses (315 μJ)

(c) Similar measurements to (b) with higher pump energy (530 μJ)

Fig.5 Experimental results of 1 550 nm pump wavelength

In Fig. 5 (a), the corresponding results for $\lambda_p = 1\ 550$ nm are presented. A similar erasing effect lasting for ~3 ps was observed, which is clear in the inset of Fig. 5(a). In addition to the erasing effect, a significant enhancement of the signal intensity at zero delay was observed. In case of p=100 mbar, the signal intensity was enhanced by one order of magnitude (red line). As to the mechanism for this enhancement, we noticed that

such an enhancement has been observed in a previous report where an 800 nm pulse was used as the main pump laser and a synchronized MIR pulse at 1 580 nm served as a control pulse[23]. This enhancement has been attributed to the fact that the 1 580 nm pulse can excite the A-B coherence by a two-photon resonance process[23]. In our current experiment, when the 800 nm and 1 550 nm pulses are synchronized in the time domain, such enhancement should also be expected since the 1 550 nm is very close to two-photon resonance occurring at 1 580 nm. While for 1 250 nm pump pulse, its photon energy is far away from A-B resonance with two photons and no such enhancement at temporal overlap was observed. To have a complete knowledge of the evolution of the emission spectrum, we present in Fig.5(b) the whole spectrum intensity as a function of the time delay. In additional to the main emission peaked at 391.4 nm, a series of relatively low intensity peaks around 388.5 nm show up, which correspond to the R branch of the B-X transition[24]. We noticed that the erasing effect and the enhancement at zero delay occur simultaneously for both the P and R branch. We also tested higher energy of the pump pulse at 530 μJ and the result is presented in Fig.5(c). In addition to the P and R branches of the B-X transition, another emission peak located at 357.8 nm appears, which corresponds to transition between the levels B ($\nu'=1$) and X ($\nu=0$)[1]. Between the optical transitions at 391.4 nm and 357.8 nm, a continuous broad band emission was observed. Recently, it has been revealed that this broadband emission origins from the optical transition that has been shifted in energy due to the dynamic Stark effect of the intense laser pulses[21,25]. Here it is observed that this broad continuum was also substantially suppressed by the delayed 800 nm control pulse.

3 Discussion

How should we understand this erasing effect of the free induction decay radiation by the delayed 800 nm femtosecond laser pulse? It is known that the 391.4 nm emission in case of MIR pump is due to the macroscopic polarization formed between the B and X states which is resonantly excited by multiple photons[21,23]. In the recent study of Yao *et al*, a theoretical description of the nitrogen ions system by the density-matrix formulism has been developed and the radiation field was described by the Maxwell equation[23]. The macroscopic polarization between the B and X states corresponds to the off-diagonal term ρ_{BX} of the density matrix [23].

Here, we speculate that the suppression of the radiation at 391.4 nm by the 800 nm pulse could be due to two reasons. The first reason lies in the fact that the control pulse also ionizes the neutral nitrogen molecules. Due to its limited pulse energy and low laser intensity, we expect that most of the produced nitrogen ions is populated in the electronic fundamental X state of the ions, and the additional population to the upper A and B levels is neglectable. Since the 800 nm control pulse is far away from the B-X resonance, the increased population in the X state due to the control pulse is incoherently added to the pre-formed nitrogen ions system by the MIR pump pulse, which can lead to decrease of the B-X coherence due to incoherent mixing. Second, we noticed that the 800 nm field is near resonant with the X($\nu=0$) and A ($\nu=2$) transition. This field, in principle, could excite the ρ_{AX}, ρ_{BA} and ρ_{BX} coherences in such way that the latter is depleted, leading to radiation suppression. We modelled this effect with our 1D Maxwell-Bloch code DeepOne[26-27].

$$\frac{\partial E_2}{\partial t}+c\frac{\partial E_2}{\partial z}=\frac{\mathrm{i}\omega_{XA}}{2\varepsilon_0}P_{AX} \tag{1}$$

$$\frac{\partial E_3}{\partial t}+c\frac{\partial E_3}{\partial z}=\frac{\mathrm{i}\omega_{BX}}{2\varepsilon_0}P_{BX} \tag{2}$$

$$\frac{\partial P_{BX}}{\partial t}=N\mu_{BX}\frac{\partial \rho_{BX}}{\partial t}=\mathrm{i}\frac{\mu_{BX}^2E_3}{\hbar}(N_X-N_B)-\mathrm{i}\frac{\mu_{AX}\mu_{BX}E_2}{\hbar}N\rho_{BA}-\frac{P_{BX}}{T_{BX}} \tag{3}$$

$$\frac{\partial P_{AX}}{\partial t}=N\mu_{AX}\frac{\partial \rho_{AX}}{\partial t}=\mathrm{i}\frac{\mu_{AX}^2E_2}{\hbar}(N_X-N_A)-\mathrm{i}\frac{\mu_{AX}\mu_{BX}E_3}{\hbar}N\rho_{AB}-\frac{P_{AX}}{T_{AX}} \tag{4}$$

$$\frac{\partial \rho_{BA}}{\partial t}=\mathrm{i}\frac{\mu_{BX}E_3}{\hbar}\rho_{XA}-\mathrm{i}\frac{\mu_{XA}E_2^*}{\hbar}\rho_{BX}-\frac{\rho_{BA}}{T_{BA}} \tag{5}$$

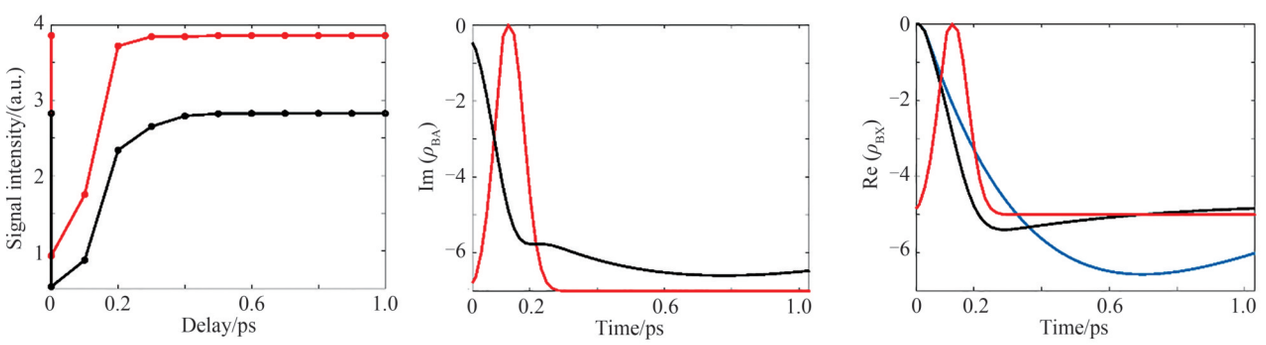
$$\frac{\partial N_{\mathrm{B}}}{\partial t}=-\frac{N_{\mathrm{B}}}{T_{\mathrm{B}}}+\frac{1}{2\hbar}\mathrm{Im}\left(E_3^{*}P_{\mathrm{BX}}\right) \tag{6}$$

$$\frac{\partial N_{\mathrm{A}}}{\partial t}=-\frac{N_{\mathrm{A}}}{T_{\mathrm{A}}}+\frac{1}{2\hbar}\mathrm{Im}\left(E_2^{*}P_{\mathrm{AX}}\right) \tag{7}$$

$$\frac{\partial N_{\mathrm{X}}}{\partial t}=\frac{N_{\mathrm{B}}}{T_{\mathrm{B}}}+\frac{N_{\mathrm{A}}}{T_{\mathrm{A}}}-\frac{1}{2\hbar}\mathrm{Im}\left(E_3^{*}P_{\mathrm{BX}}\right)-\frac{1}{2\hbar}\mathrm{Im}\left(E_2^{*}P_{\mathrm{AX}}\right) \tag{8}$$

In these equations E_2, ω_{XA}, and E_3, ω_{BX} are respectively the electric field and frequency of the λ = 800 nm and λ = 391 nm radiation fields. P_{ij}, ρ_{ij} and μ_{BX} are respectively the macroscopic polarization density, coherences and electric dipoles between X, A and B levels of the ionized nitrogen molecules. N is the density of ionized nitrogen molecules and N_i the population of each level. Finally, $T_{ij}=2\times11^{11}$ s^{-1} are depolarization characteristic times and $T_i=500\times10^{-12}$ s are the radiative lifetimes of the levels.

To model the experiment we asume a 1 mm nitrogen plasma, at 50 mbar pressure and 10% ionization with an initial B-X coherence, (ρ_{BX}=0.001) We inject a 100 μJ, 100 fs, IR pulse in resonance with the X-A transition (λ~800 nm) at different delays and compare the total energy of the UV radiation (λ=391 nm) emitted due to the initial ρ_{BX} coherence.



(a) UV intensity as a function of the time delay for 30 mbar (black) and 50 mbar (red)

(b) Temporal dynamics of the imaginary part of ρ_{BA} (black) induced by a delayed (100 fs) IR pulse (red)

(c) Temporal dynamics of the real part of ρ_{BX} (black) induced by a delayed (100 fs) IR pulse (red). The unperturbed ρ_{BX} (blue) is also shown.

Fig.6 Results of the 1D Maxwell-Bloch modelling

The results of our 1D Maxwell-Bloch modelling are shown in Fig. 6. The dependence of the intensity of the emitted 391 nm radiation with the delay of the 800 nm control pulse is shown in Fig. 6(a) for two different pressures, 30 mbar (black) and 50 mbar (red). As expected, a sudden drop in the intensity and posterior recovery when the delay of the IR control pulse is increased is apparent in Fig. 6(a). This is explained as follows. The IR control pulse and the initial ρ_{BX} coherence strongly induces a negative imaginary part on the ρ_{BA} coherence, as shown in Fig. 6(b). The ρ_{BA} coherence then interacts with the IR control pulse, depleting the ρ_{BX} coherence, as depicted in Fig. 6(c). Since most of the emission induced by the ρ_{BX} coherence takes place during less than one picosecond, the intensity recovers when the delay increases.

In view of these results, we can affirm that the two aforementioned mechanisms (further ionization by the IR control pulse and depletion of the ρ_{BX} coherence by the interaction of the IR control pulse and the ρ_{BA} coherence) play a role in the dynamics of the system.

4 Conclusion

We have demonstrated that the FID emission of nitrogen ions pumped by femtosecond MIR pulses can be substantially suppressed by a subsequent 800 nm control pulse. This erasing effect occurs universally for different pump laser wavelength, gas pressure and control pulse energy. In addition to the erasing effect, a significant enhancement of the spectral signal was also found for λ_{p} = 1 550 nm, where two-photon resonance

between the B and A levels occurs. Based on the numerical simulation of the density matrix and Maxwell equation, we attribute this erasing effect to the further photoionization by the 800 nm pulse and the change of coherence between the B and X states of the nitrogen ions. Specifically, the 800 nm pulses directly leads to the change of coherence between the B and A states (ρ_{BA}). The further coupling of the ρ_{BA} with the 800 nm pulse results in the reduction of the coherence of the B and X states (ρ_{BX}), which gives rise to the corresponding suppression of the 391.4 nm radiation. This erasing effect provides a simple method for measurement of the temporal duration of the FID emission, which is particularly useful when the weak FID emission overlaps with the harmonics of the pump laser in the spectral domain.

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Erasing of the Free Induction Decay by Incoherent Ionization Injection

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Abstract: Strong field physics is an important frontier of current physics research. In recent years, the rapid development of ultrafast laser technology has enabled researchers to obtain laser sources with shorter pulse width, higher pulse energy, and wider tunable range. With the advancing of the research on the interaction between strong laser and matter from the traditional perturbation regime to the non-perturbative region, rich strong field physical phenomena such as tunneling ionization and high-order harmonics have been observed. When the high-intensity femtosecond laser propagates in in a transparent medium, it can form a light-plasma filament due to nonlinear optical effects including Kerr effect and plasma generation. In particular, the study of "air lasing" radiation produced by interaction of intense femtosecond laser pulses with air molecules has attracted many attentions in recent 10 years. Air lasing has the characteristics of bidirectional emission, high intensity, high coherence and remote generation, and holds potential application in the field of optical remote sensing. Traditional optical remote sensing collects scattered signals or fluorescent signals of lasers on the atmospheric targets. These optical signals do not have directionality, which poses challenges to the sensitivity of ground collection devices and limits detection accuracy and sensitivity. As a new concept light source, air lasing is expected to greatly improve the signal strength of optical remote detection due to its capacity of emitting coherent beams from the remote atmosphere to the ground. In recent years, several important progress have been achieved as to the understanding of the air lasing signal generated by the interaction of strong-field laser and the most important component of air, nitrogen. Regarding the underlying mechanism of the coherent emission of neutral nitrogen molecule, it is now accepted that the emission at 337.4 nm is Amplified Spontaneous Emission (ASE). In contrast, the lasing mechanism of N_2^+ is much more complex and is still hotly debated. Part of the debate stems from the mysterious fact that coherent emissions at 391.4 and 427.8 nm can always be observed regardless of the pump laser wavelength at 400 nm, 800 nm, 1 100 to 1 900 nm, or even 3 900 nm. Very recently, when pump lasers in the mid-infrared regime were used as drive pulses, the nature of the 391.4 and 427.8 nm emissions was attributed to a Free Induction Decay (FID) signal. In this study, we report for the first time the erasure effect of the FID signal pumped of nitrogen ions under an 800 nm control pulse. This effect provides a unique tool to measure the time-domain pulse width of the weak free induction decay signals, which is especially useful in the case where the FID radiation and the harmonics of the pump pulse overlapping in the frequency domain. In our study, by pumping nitrogen ions with strong-field mid-infrared femtosecond pulses (1 250/1 550 nm), the 391.4 nm radiation corresponding to state $B^2\sum_u^+(\nu'=0)$ and state $X^2\sum_g^+(\nu=0)$ transitions was observed in the forward direction. When pumped by

1 250 nm femtosecond pulses, the coherent 391.4 nm signal overlaps with the third harmonic in the spectral domain, while the spectrum of the 391.4 nm signal is clean when nitrogen gas was pumped by 1 550 nm femtosecond pulses since the the 391.4 nm wavelength deviates from the third and fifth harmonics. In our experiment, time-resolved measurement was performed by injection of an 800 nm control pulse. It was found that the subsequent 800 nm control pulse would have a significant suppression effect on the FID signal generated by the 1 250/1 550 nm pump laser. The suppression of the FID signal lasts approximately 3 ps. For 1 250 nm pump pulse, it was found that the FID signal was suppressed by about 75% for control pulse energies of 200 μJ and 300 μJ. While for the 100 μJ pulse, the FID signal is less suppressed. For 1 500 nm pump pulse, in addition to a similar erasure effect, at zero delay we observed a significant enhancement of the signal. This enhancement is attributed to the fact that the 1 550 nm pulse can excite the A-B coherence through a two-photon resonance process. While for the 1 250 nm pump pulse, its photon energy is far away from the A-B resonance with two photons, so no such enhancement is observed at the temporal overlap. Simultaneously, a similar erasing effect can be observed for 357.8 nm radiation corresponding to levels B ($\nu'=1$) and X ($\nu=0$) transitions when pumped by high-energy (530 μJ) 1 550 nm pulses. Based on numerical simulations, we attribute the mechanism of this erasure effect to the photoionization and reduction of the coherence between the B and X states due to the 800 nm control pulse. It was found that the intial B-X coherence couples with the 800 nm control pulse and leads to the change of B-A coherence, which in turn couples with the 800 nm pulse and results in reduction of the B-X coherence.

Key words: Air lasing; Ultrafast spectroscopy; Free induction decay; Femtosecond pulses

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