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Optimally enhanced optical emission in laser-induced air plasma by femtosecond double-pulse

Anmin Chen,[1,2] Suyu Li,[1] Shuchang Li,[1] Yuanfei Jiang,[1] Junfeng Shao,[3] Tingfeng Wang,[3] Xuri Huang,[2] Mingxing Jin,[1,3,a)] and Dajun Ding[1]
[1]*Institute of Atomic and Molecular Physics, Jilin University, Changchun 130012, China*
[2]*Institute of Theoretical Chemistry, State Key Laboratory of Theoretical and Computational Chemistry, Jilin University, Changchun 130012, China*
[3]*State Key Laboratory of Laser Interaction with Matter, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun 130033, China*

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In laser-induced breakdown spectroscopy, a femtosecond double-pulse laser was used to induce air plasma. The plasma spectroscopy was observed to lead to significant increase of the intensity and reproducibility of the optical emission signal compared to femtosecond single-pulse laser. In particular, the optical emission intensity can be optimized by adjusting the delay time of femtosecond double-pulse. An appropriate pulse-to-pulse delay was selected, that was typically about 50 ps. This effect can be especially advantageous in the context of femtosecond laser-induced breakdown spectroscopy, plasma channel, and so on. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4825346]

I. INTRODUCTION

Over the past many years, femtosecond laser-induced air plasma has received increased attention due to its importance in many applications, such as: plasma channel generation,[1] supercontinuum generation,[2] terahertz emission,[3] environmental monitoring,[4] and so on. Plasma channel produced by lasers in atmospheric air or some other gases are of great interest for many fundamental problems and technical applications.[5] Using air as an emitting medium to generate terahertz waves can be used for standoff distance terahertz wave sensing and imaging, which also attract much attention in recent years.[6]

Researchers are paying more attention to the intensity and lifetime of femtosecond laser-induced air plasma as the key parameters in the current experiments. One of the possible ways is to add supporting laser pulses following the lead femtosecond laser pulse which generates the plasma channel.[7–9] Prolonging of the channel lifetime by adding a delayed long laser pulse has been investigated and practiced in filament triggered HV discharge experiment.[10–12] Recently, femtosecond laser pulse sequence had been used in the experiment to further prolong the lifetime of the plasma channel.[13]

In this paper, femtosecond double-pulse was introduced into the study of laser-induced air plasma. The study of the mechanisms would lead a further improvement to the applications of femtosecond double-pulse. The produced optical emission was observed in the delay time range from −200 ps to 300 ps. Femtosecond double-pulse laser air ionization will benefit some key technological applications.[6,14,15] Compared with the investigation of air plasma generated by the femtosecond single-pulse laser[10] and the femtosecond double-pulse at the fixed time intervals,[14–16] the higher optical emission intensity of air plasma can be obtained by adjusting the delay time of femtosecond double-pulse.

a)Electronic mail: mxjin@jlu.edu.cn.

II. EXPERIMENTAL SETUP

The experimental setup for femtosecond double-pulse laser-induced plasma spectroscopy measurement is shown in Fig. 1. The laser system is a regenerative amplified Ti: Sapphire laser (Spectra Physics Tsunami oscillator and Spitfire amplifier). The full-width at the half maximum (FWHM) is 120 fs, the wavelength is 800 nm, and the repetition rate is 1 kHz. The maximum energy of the output femtosecond single pulse is about 300 μJ. The individual pulse is splitted into two sub-pulses by a beam splitter. Using a computer-controlled translation stage (Physik instrumente, M-505), the delay time may be changed from −300 to 400 ps. By the combination of a Glan laser polarizer and a half-wave plate, the energy of one sub-pulse can be attenuated to the desired value (86 μJ and 60 μJ). At the same time, we use this sub-pulse as main pulse and a zero reference time point during the whole experiment. The energy of another sub-pulse is fixed to 26 μJ. The subpulses are directed by a beam splitter into a microscope objective ($10 \times \mathrm{NA} = 0.25$). The laser-induced air plasma spectroscopy perpendicular to the laser beam is collected by lens (BK7), focused into a fiber, and detected with the spectrometer (Avantes, AvaSpec-FAST).

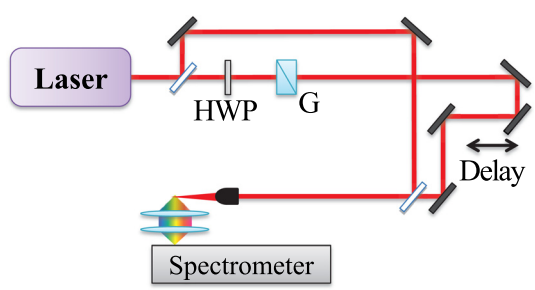

FIG. 1. Experimental setup for spectroscopy measurement of femtosecond double-pulse laser air plasma. HWP is half-wave plate. G is Glan laser polarizer.

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All experiments are performed in air at atmospheric pressure. The each spectroscopy is an average of typically 200 shots.

III. RESULTS AND DISCUSSION

At the beginning of the experiment, the sub-pulse of fixed energy (26 μJ) was blocked. When the energy of main pulse was 60 μJ, the spectrometer can detect weaker signal. The energy of main pulse was adjusted to 50 μJ, the signal of spectroscopy cannot be almost observed by the spectrometer. The signal of spectroscopy shown in the spectrometer was only the noise. The air plasma cannot be produced by the laser energy of 26 μJ.

The time-integrated emission spectra of the femtosecond double-pulse laser-induced air plasmas in a spectral range of 300–700 nm were recorded with the delay time of femtosecond double-pulse, as shown in Fig. 2. The total energy of double-pulse was 112 μJ (86 + 26 μJ). As can be seen in the figure, the spectral intensity of air plasma was not found to depend on the delay time of double-pulse in −200−0 ps range. Since the energy of 26 μJ just was lower than the multiphoton-ionization threshold. The second pulse was too early, as the air plasma was still not produced. It was equivalent to the single pulse ionized air. We randomly selected the spectroscopy of the delay time of −50 ps, this spectroscopy was considered as the average emission intensity in −200–0 ps range. In the range of 0–350 ps, the spectral intensity was significantly different compared with the range of −200–0 ps. Fig. 3 shown emission intensity at the delay time of −50 ps, 20 ps, 40 ps, and 300 ps. The intensity of air plasma was significantly enhanced when the main pulse was followed by a smaller energy pulse. In Fig. 3(a), the enhancement factor was about 5 at the delay time of 50 ps.

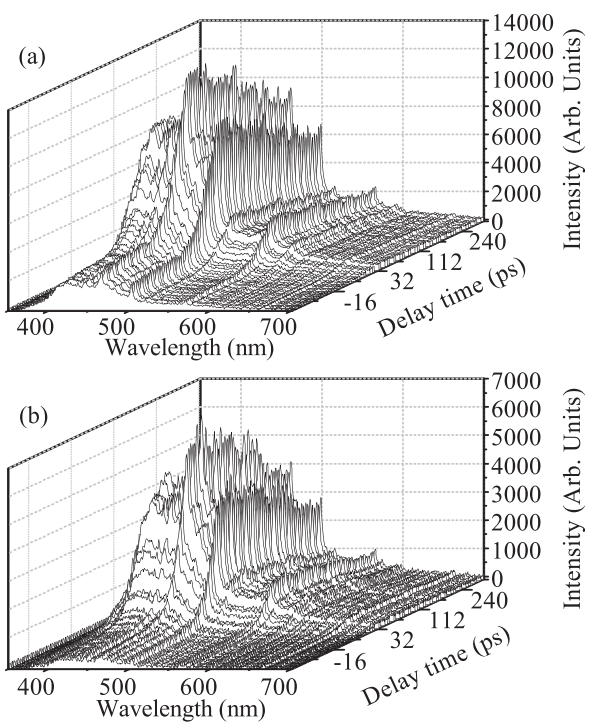

FIG. 2. The distribution of spectral intensity with the delay time of double-pulse laser. The energy of femtosecond double-pulse laser is 86 + 26 μJ (a), and 60 + 26 μJ (b).

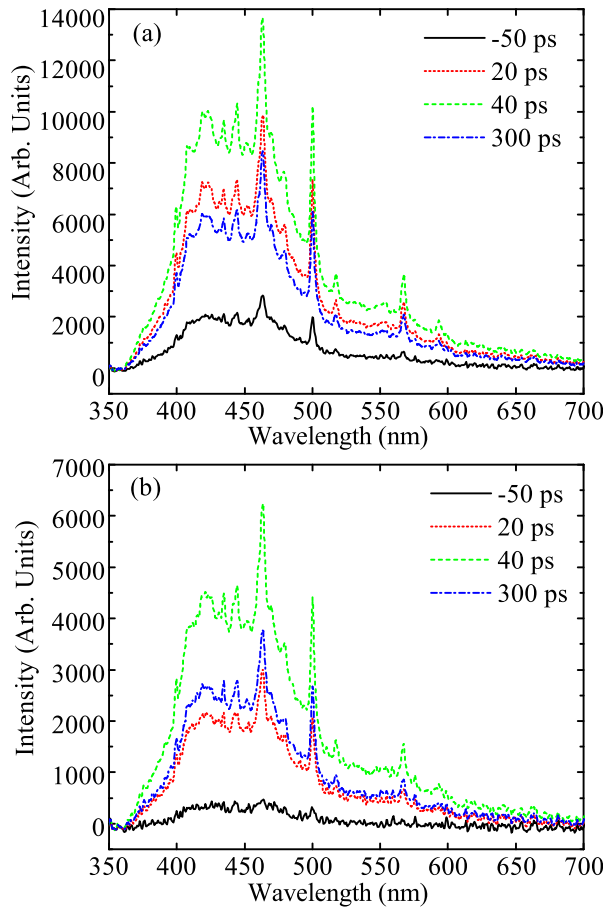

FIG. 3. The spectral intensity at the delay time of −50 ps, 20 ps, 40 ps, and 300 ps. The energy of femtosecond double-pulse laser: (a) 86 + 26 μJ; (b) 60 + 26 μJ.

The plasma consists of different species: electrons, ions, and neutral atoms. Due to this, the absorption of the radiation inside the plasma occurs by different mechanisms. There are two main mechanisms by which the radiation is absorbed by the plasma: inverse Bremsstrahlung (IB) and multiphoton absorption (MPA) processes.[17] In this experiment, the interaction of air and first pulse laser was described by MPA mechanism. Under this mechanism, a large number of free electrons were produced by the MPA. The second pulse laser was absorbed of by free electrons in the plasma, the dominant mechanism is IB. The absorption efficiency of IB is much higher than the absorption efficiency of MPA.

The emission intensity is proportional to the transition probability and the concentration of the excited atoms. Since the transition probability for the emission is considered to be unchanged, the enhancement effect is attributed to the increase in the concentration of the excited atoms caused by double-pulse laser-induced plasmas.[18] When the plasma was generated, the main pulse laser was employed to pre-ionize air, the density and temperature of plasma was increased to a very high level.[19] As a result, the region of produced plasma was not transparent. It increased the absorption of the laser in this region, causing the increase in the plasma density and temperature at this region.[20] Due to this absorbed effect for plasma, the emission intensity from the plasma was enhanced.

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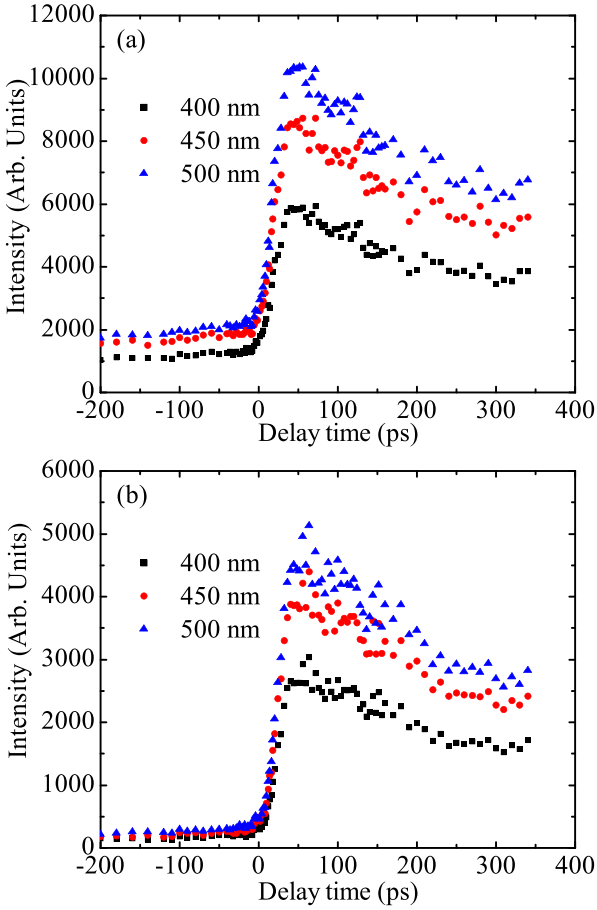

FIG. 4. The spectral intensity vs. the delay time of double-pulse laser at the different wavelength. The energy of femtosecond double-pulse laser is: (a) 86 + 26 μJ; (b) 60 + 26 μJ.

In order to understand the details of the double-pulse enhancement effect, Fig. 4 shown the influence of the delay time of double-pulse on the intensity of the plasma emission signal. The wavelength of 400 nm and 500 nm was line spectrum, 450 nm was continuous spectrum. According to this plot, three main regimes were distinguished. In the range of −200–0 ps, we noticed that the enhancement of emission intensity was not sensitive to the delay time of double-pulse. At this moment, the main pulses became the second pulse, the emission intensity of the produced plasma was equivalent to the emission intensity of single-pulse laser-induced plasma. And the whole laser energy equalled to the energy of the main pulse. Because, the energy of 26 μJ was lower than the threshold of air ionization. A transition regime was clearly observed in the range of 0 to 50 ps. Within this interval the emission intensity of plasma increased steadily compared to the range of −200–0 ps and a maximum value was reached at approximate 50 ps. Further increasing the delay time, the intensity began to drop slowly, and then kept constant. Fig. 5 contrasted the variation of the emission intensity for 112 μJ (86 + 26 μJ) and 86 μJ (60 + 26 μJ) at the wavelength of 450 nm. The straight line in the figure was the average intensity of single-pulse of 86 μJ produced plasma. Compared double-pulse of 60 + 26 μJ and single-pulse of 86 μJ, the maximum enhancement factor was around 2 at the delay time of 50 ps. With increased the delay time, the factor

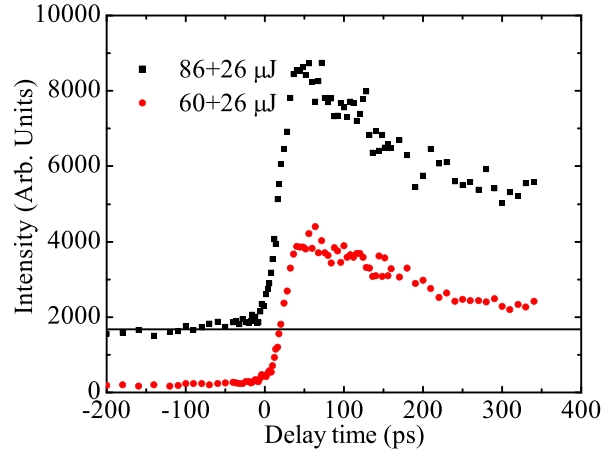

FIG. 5. The spectral intensity vs. the delay time of double-pulse laser at the same wavelength with the different energy of femtosecond double-pulse. The wavelength is 450 nm.

of enhancement reduced, and gradually closed to the emission intensity of 86 μJ.

During the multiphoton ionization air, the evolution of the electron density is described by the Drude model[21,22]

$$\frac{\partial n_e}{\partial t} = \frac{\beta^{(K)}|E|^{2K}}{K\hbar\omega} + \frac{1}{n_b^2}\frac{\sigma}{E_g}|E|^2 n_e - a n_b^2, \tag{1}$$

where n_e is the electron number density, t is the decay time, $\beta^{(K)}$ is the coefficient of K-photon ionization, E is the laser electric field, $\hbar$ is the Planck constant, ω is the laser frequency, $n_b \approx 1$ is the index of refraction of air, $E_g = 12.2$ eV is the ionization energy, $a = 5.0 \times 10^{-13}$ m^3/s is recombination rate. The cross section can be calculated as[23]

$$\sigma = \frac{ke^2\tau}{\omega m_e \epsilon_0}\frac{1}{1+\omega^2\tau^2}, \tag{2}$$

where k is the wave number, e is the electron charge, τ is the mean free time between momentum transfer collision,[19] m_e is the electron mass, ϵ_0 is the dielectric constant of vacuum. Using this simple model, we presented the rough calculated result for the electron density of femtosecond laser multiphoton ionization in air at atmospheric pressure.

The evolution of electron density with the decay time in femtosecond laser ionization region was shown in Fig. 6. From this plot, the electron density rose rapidly and reached at maximum by multiphoton ionization processes. The higher electron density kept about 10 ps. Subsequently, the electron density decreased with time due to the effect of electron recombination in the ionization region. Obviously, based on the IB processes, the absorption of light should be optimal for the second pulse at the range of 0–10 ps. The emission intensity of the produced plasma spectroscopy should be higher, too. However, at about 50 ps, the plasma spectroscopy obtained the maximum emission intensity by IB processes of the second pulse (Fig. 4). So, we had to explain this phenomenon by other means.

The propagation of femtosecond pulses had Gaussian radial intensity distribution behind a focusing lens.[24] Near the

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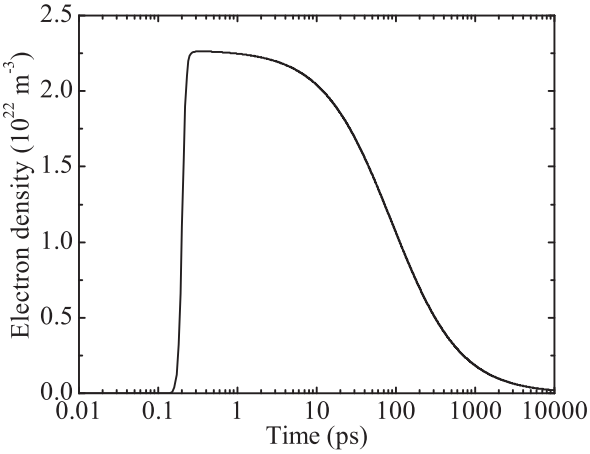

FIG. 6. The evolution of electron density with the decay time in femtosecond laser ionization region.

threshold of air photoionization by 800 nm femtosecond laser, the air ionization was occurred in the center region of laser Gaussian intensity. The original spatial region of air ionization was shown in Fig. 7(a). Most of light of the second pulse is transmitted through the region. A part of the Gaussian center light may be absorbed by IB processes. With increase of the delay time, the ionization region quickly increased (as seen in Fig. 7(b)). At the same time, the laser absorption also increased by free electron in the air plasma. And, the emission intensity of air plasma quickly increased. Until the region of air plasma was equal to the area of laser focal point. At this time (as seen in Fig. 7(c)), the maximum was reached for the light absorption. The emission intensity of air plasma obtained the maximum. In the range of the delay time, the produced plasma had always the higher electron density (as seen in Fig. 6). Continue to increase the delay time, based on the expansion of the plasma (Fig. 7) and the electron recombination (Fig. 6), the density of free electron gradually began to decrease. The light absorption of the plasma and the intensity of spectroscopy decreased.

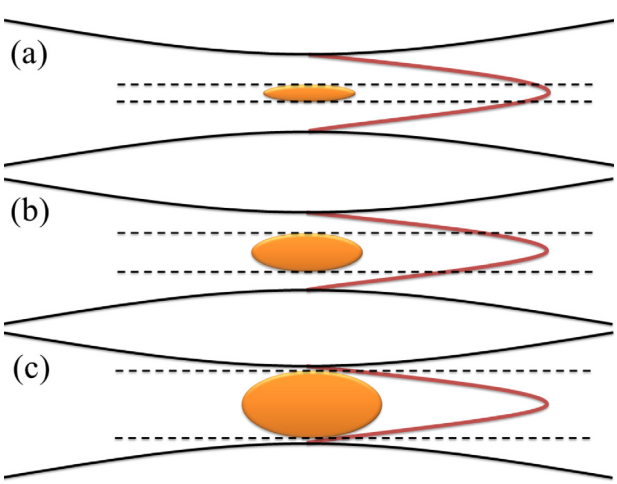

FIG. 7. The spatial evolution of plasma in femtosecond laser focusing air breakdown: (a) original region of air ionization, (b) and (c): the expansion of air plasma ionization region.

IV. CONCLUSIONS

In conclusion, with ambient air as the medium we demonstrated enhancement of emission by the femtosecond double-pulse laser-induced air plasma. The amplitude of enhancement increased following the delay time of the double-pulse. Enhancement lasting up to 50 ps was observed. By recording the dependence of spectral intensity on the delay time of double-pulse, the enhancement of the optical emission can be optimized. This method can be a tool for environmental monitoring, element analysis, and remote sensing. It may also be used to enhance the intensity and lifetime of plasma channel generation and emission of terahertz radiation.

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