CHIN.PHYS.LETT. Vol. 16, No. 8 (1999) 580

Plasma Generation in Air by Intense Laser Pulses with Various Pulse Durations *

QIN Yuan-dong(秦元东), YANG Hong(杨宏), WANG Shu-feng(王树峰), ZHU Chang-jun(朱长军),
GONG Qi-huang(龚旗煌)**
Department of Physics, Mesoscopic Physics Laboratory, Peking University, Beijing 100871

(Received 16 March 1999)

We have studied the plasma generation through collecting the charged particles produced in laser-induced breakdown in atmospheric air. This breakdown is produced by ultrashort laser pulses with various durations ranging from 200 fs to 2 ps at a fixed pulse energy of 8 mJ. We found that the plasma yield increases first with the increment of pulse duration and then reaches a plateau at a duration longer than 500 fs. The generated plasma temperature is estimated to be higher than 5×10⁵ K.

PACS: 52. 50. Jm, 42. 65. Re, 33. 80. Rv

Intense light–matter interaction is attracting much attention after the appearance of high power laser.[1] At a high intensity of 10^{11} W/cm^2, almost all the atoms and molecules can be easily multiphoton-ionized,[2] The air is fully ionized when the laser intensity reaches about 10^{13} W/cm^2 (pulse duration about 10 ns).[3] When high power laser interacted with dense gas medium or condensed matter, laser-induced breakdown (LIB) which refers to the plasma formation through multiphoton and cascade ionization usually occures.[4] In such a process, the extremely high intensity of ultrashort pulses first leads to a multiphoton absorption in the material and it provides initial charged particles for LIB.[5] The free electrons and ions absorb energy from the electromagnetic field of the laser radiation by inverse bremsstrahlung and they are accelerated.[6] The subsequent avalanche-like multiplication (cascade ionization) of free carriers finally leads to LIB. And LIB is usually accompanied with visible flash or spark, which results from plasma emission. It has been observed in solids,[7,8] liquids,[9] and gases.[10]

The LIB has been studied in many fields. The rapid formation of plasma and subsequent blocking to input laser pulse,[11] can act as a high-speed optical switch. The LIB has also achieved widespread clinical use in various ophthalmic laser surgeries.[12,13] Recently, researchers have began to study harmonic generation (an efficiency of third harmonic generation as high as 1.7×10^{-3} was reported in femtosecond LIB in atmospheric air[14]), white continuum generation[15,16] and chemical product[17] in LIB. In this letter, we investigated experimentally the relation between the plasma generation and pulse duration in pico- and subpico-second range by directly measuring the amount of charged particles. These results might be of benefit to the further study of subpicosecond pulse with matter.

The experimental setup for LIB in air was based on the Spectra-Physics Ti:Sapphire femtosecond system with a regenerative Chirp-Pulse-Amplifier (TSA-10). The laser system generated 800 nm light pulse with a repetition rate of 10 Hz. The maximum energy was up to 13 mJ per pulse with fluctuation less than 5%. We obtained different pulse durations by adjusting the compressor. In the experiment, the pulse duration was changed from 216 fs to 2.085 ps at a fixed pulse energy of 8 mJ. The output laser beam was splitted into two parts, one is strongly attenuated and feeded into an autocorrelator to measure the pulse duration, the other beam (~8mJ/pulse) was focused into a spot of about 61 μm in diameter (about 3 times of diffraction limit) with a 12 cm focal lens. The maximum irradiance in the focus was estimated to be 2.2×10^{15} W/cm^2.

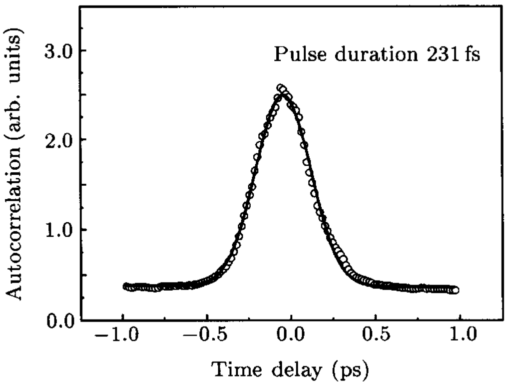

Fig. 1. Autocorrelation signal, open circles for the experimental data and the solid line for the fitted curve with a Gaussian expression.

The autocorrelator gave out the pulse duration, as shown in Fig. 1. The focused beam induced a plasma of optical breakdown in the atmospheric air, which was indicated by the appearance of a spark. Two metal plates enveloping the plasma were deliberately designed to act as electrodes to collect the generated charged particles. The distance between the electrodes was about 1 mm with the applied dc voltage 36 V. This applied dc electrostatic field helped the generated charged particles move to the electrodes. The current signals at various pulse durations were collected and demonstrated on an oscilloscope.

---

*Supported by the National Climbing Project of China, and the National Natural Science Foundation of China under Grant Nos. 19884001 and 19525412.

**To whom the correspondence should be addressed.

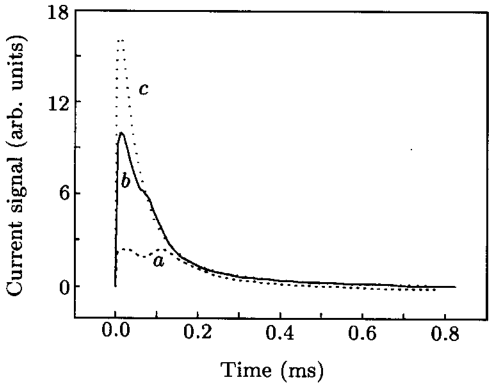

Fig. 2. Current signals at different pulse duration τ = 231 fs (a), 334 fs (b), and 934 fs (c).

Figure 2 is the typical current signals at various pulse durations. The current signal has two distinctive peaks with amplitudes increasing with the increment of pulse duration. Another remarkable feature is that the second peak was forward-shifted as the pulse duration was enlarged. Two peaks emerged into one when the pulse duration reached 934 fs. We predict that the first peak is probably the contribution from electrons and the second one from ions. Although they experienced a lot of collisions with other electrons, ions and molecules, which are inevitable in LIB in air, the most of the electrons still reached the electrode earlier than the ions because of their much smaller masses. The above prediction was verified by the obvious shift of the second peak when we moved the electrodes in the direction perpendicular to the focused laser beam.

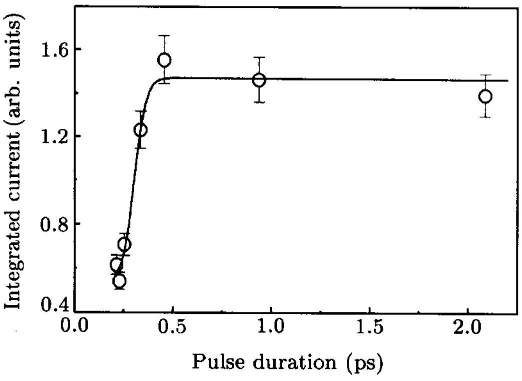

Fig. 3. Integrated current signals at various pulse durations from 216 fs to 2.085 ps. The solid line is used to guide the eyes.

Figure 3 shows the dependence of integrated current signal on pulse duration. The integrated current signal roughly reflects the number of the charged particles, where we assume a constant electron–ion recombination rate. From Fig. 3, we infer that the plasma yield in LIB increases rapidly with the increment of pulse duration then reaches a plateau as the pulse duration longer than 500 fs. We should point out that this particular behavior of plasma production was definitely not originated from the so-called "stabilization", because the laser field we used was not enough to induce stabilization (usually$>10^{16}$ W/cm^2). This was also confirmed with the experimental result at a short duration (about 300 fs); we found that the plasma yield increased with increasing pulse intensity, which is contrary to the stabilization theory. As it is well-known that the interaction between laser pulse and the generated plasma will shield the input pulse energy. In a recent pump-probe experiment,[18] we found an energy loss up to 50% for the probe beam when LIB was induced by the intense pump beam at pulse duration of 280 fs. The main mechanism of plasma shielding is inverse bremsstrahlung absorption of photons during collisions between electrons and heavy particles (molecules or ions). The energy absorbed by the plasma generated a very high temperature ($>$ 5000 K) within the plasma[19] and resulted in a rapid expansion. The higher energy absorption would lead to a higher plasma yield. The higher plasma yield would promote the energy absorption in return.

The following rate equation was used to describe the plasma generation:[4,20]

$$\frac{\partial\rho}{\partial t} = \eta\rho - g\rho + (\frac{\partial\rho}{\partial t})_m, \tag{1}$$

where ρ is the electron density. The first term in the right-hand side represents cascade ionization and the cascade ionization rate η is the probability per unit time during which a free electron will have an ionizing collision with a molecule. We have the form:

$$\eta = \left(\frac{e^2|E|^2}{m} - \frac{2m\varepsilon_{\text{av}}\omega^2}{M}\right)\frac{\tau' E_{\text{ion}}}{1+\omega^2\tau^2}, \tag{2}$$

where E is the laser field with a frequency ω, τ' the momentum transfer collision time which is reversely proportional to air pressure, ε_{av} the average electron energy, M the atomic(or molecular) mass, E_{ion} the binding energy of a bound electron, g represents free electron loss due to recombination, trapping, and diffusion out of the focal volume of the beam. The third term in the right-hand side of Eq. (1) represents carrier generation by multiphoton ionization. The expression of η indicates cascade ionization is only meaningful during laser pulse.

Multiphoton ionization is a nonlinear optical process which becomes significant only at high irradiances and for wavelengths in the near-infared or shorter.[20] It is potentially much faster than the buildup of an electron cascade and therefore occurs even for the shortest laser pulse. In an ultrashort pulse regime the pulse may be so short that cascade cannot build up during the pulse.[20] The experimental data of multiphoton ionization rate is not available for air LIB. However, as a reference, Ne is ionized to Ne$^+$ with a rate of about 10^{15} s^{-1} at a laser intensity of 10^{15} W/cm^2 as a calculation result.[21]

Now, we take a rough estimation of the collision time τ' (between an electron and an air molecule) during cascade ionization. Because the plasma cools on a subnanosecond time scale, the ions usually do not have time to equilibrate with the electrons, so nearly all of the energy is coupled into the electrons during the pulse we studied.[22] For a plasma density of about $10^{19}\,\mathrm{cm}^{-3}$ (as suggested in Ref. 23) and a plasma volume of about $\pi \times (0.003)^2 \times 0.5 = 14.13 \times 10^{-6}\,\mathrm{cm}^3$ (the volume has a radius of 0.003 cm and a length of 0.5 cm), we assume an input pulse energy absorption of 20–50% by the plasma. The calculated plasma temperature is 547025–1367563 K, and the average collision time is 50–80 fs. So, even for a few percent of air being multiphoton ionized at the early stage (in Ref. 24, a plasma density of $0.4\times10^{17}\mathrm{cm}^{-3}$ is obtained through tunneling ionization during the 100 fs pulse with peak intensity of $6.5\times10^{13}\,\mathrm{W/cm}^2$), there need more than 5 collisions to wholly cascade-ionize the air in the focal volume. Thus, for very short pulse, it is difficult to build up the cascade ionization during the pulse because few collisions can occur. For more clarity, we made a simple simulation on the role of cascade ionization in plasma production with Eq. (1). In the simulation, we considered only a little section of plasma zone and neglected loss of pulse energy when it passed through this section. The role of cascade ionization R is defined as

$$R = 1 - \frac{\rho_{\mathrm{without}}}{\rho_{\mathrm{with}}}, \qquad (3)$$

where ρ_{without} and ρ_{with} are the electron density without and with the cascade ionization channel, respectively; both of them were sampled just behind one pulse because the multiphoton ionization and cascade ionization just stopped at that time. The calculated results as presented in Fig. 4, indicates the same trend with the experimental results.

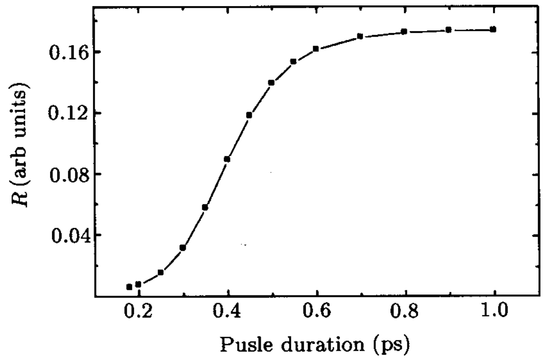

Fig. 4. Role of cascade ionization in plasma production as a function of pulse duration.

The behavior of plasma yield with various pulse durations is understood as following: The plasma formation is initiated by multiphoton ionization then cascade ionization. As no enough time for interaction between laser and molecules or produced plasma, narrower input pulse only produces less electrons and ions during the pulse duration even it has a higher peak intensity. While for a longer-duration pulse, the produced plasma has enough time to absorb laser energy to be accelerated, to expand more rapidly. The accelerated electrons will speed up plasma production through collisions with molecules or ions (which is the essence of cascade ionization). As to the plateau, we suppose that most of the air molecules in the focal volume have been ionized in a longer pulse duration and therefore lead to a saturation of plasma yield. Of course, for an even longer duration input pulse, the plasma yield will decrease because of the low peak intensity, which leads to a poor plasma production.

In summary, we studied the plasma generation at a series of pulse durations with a fixed energy. We found that the plasma yield increased with the increment of pulse duration and reached a plateau at 500 fs. From our results, the laser with a duration long than 500 fs seems favorable for harmonic generation in self-induced breakdown.

Acknowledgment: GONG Qihuang is grateful to Professor GAN Zi-zhao for his stimulated discussion and encouragement.

REFERENCES

[1] M. D. Perry and G. Mourou, Science, 264 (1994) 917.
[2] M. Luvan, G. Mainfray, C. Manus and I. Tugov, Phys. Rev. A 7 (1973) 9.
[3] I. A. Bufetov, G. A. Bufetova and V. B. Tedorov, IEEE J. Quantum Electron. 31 (1995) 1172.
[4] Y. R. Shen, *The Principle of Nonlinear Optics* (Wiley, New York, 1984).
[5] C. A. Puliafito and R. F. Steinert, IEEE J. Quantum Electron. 20 (1984) 1442.
[6] J. F. Ready, *Effects of High-Power Laser Radiation* (Academic Press, New York, 1971).
[7] D. W. Fradin, E. Yablonovitch and M. Bass, Appl. Opt. 12 (1973) 700.
[8] HE Jing-tang, ZHANG Ping, CHEN Duan-bao, LI Zuhao, ZHANG Ying, WANG Long, FENG Bao-hua, ZHANG Xiu-lan, WEI Zhi-yi, LI Zan-liang, TANG Xiao-wei and ZHANG Jie, Chin. Phys. Lett. 15 (1998) 816.
[9] M. P. Felix and A. T. Ellis, Appl. Phys. Lett. 19 (1971) 484.
[10] C. DeMichelis, IEEE J. Quantum Electron. QE-5 (1969) 188.
[11] D. C. Smith, Appl. Phys. Lett. 19 (1971) 405.
[12] J. M. Krauss and C. A. Puliafito, *Laser Surgery and Medicine: Principles and Practice*, edited by C. A. Puliafito (Wiley-Liss, New York, 1996).
[13] R. F. Steiner and C. A. Puliafito, *The Nd: YAG Laser in Ophthamology: Principles and Clinical Applications of Photodisruption* (Saunders, Philadelphia, PA, 1985).
[14] A. B. Fedotov, N. I. Koroteev, M. M. T. Loy, X. Xiao and A. M. Zheltikov, Opt. Commun. 133 (1997) 587.
[15] A. Borghese and S. S. Merda, Appl. Opt. 37 (1998) 3977.
[16] H. Nishioka, W. Odajima, K. Ueda and H. Takuma, Opt. Lett. 20 (1995) 2505.
[17] S. L. Chin and S. Lagace, Appl. Opt. 35 (1996) 907.
[18] to be submitted
[19] D. X. Hammer, E. D. Jansen, M. Frenz, G. D. Noojin, R. J. Thomas, J. Noack, A. Vogel, B. A. Rockwell and A. J. Welch, Appl. Opt. 36 (1997) 5630.
[20] P. K. Kennedy, IEEE J. Quantum Electron. 31 (1995) 2241.
[21] S. C. Rae and K. Burnett, Phys. Rev. A 46 (1992) 2077.
[22] W. L. Kruer, *The Physics of Laser Plasma Interactions* (Addison-Wesley, Redwood City, 1988).
[23] T. Auguste, P. Monot, G. Mainfray and C. Manus, Laser Phys. 4 (1994) 52.
[24] A. Braun, G. Korn, X. Liu, D. Du, J. Squier and G. Mourou, Opt. Lett. 20 (1995) 73.