

# Optical ionization evolution effect on photocurrent produced from two-color femtosecond laser pulses

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**Abstract** When laser intensity varies, the ionization induced by optical field can be described by multiphoton ionization (MPI) in low regime and tunneling ionization (TI) in high regime. An empirical formula was used to fit the ionization happened between these two limitation ionization processes. Based on this, ionization rate and photocurrent induced by two-color femtosecond laser pulses interaction with air-plasma were investigated numerically. It was found that they have different relations with the laser intensity.

**Keywords** multiphoton ionization (MPI), tunneling ionization (TI), photocurrent

## 1 Introduction

Terahertz (THz) wave generated from two-color (fundamental pulses and its second harmonic) femtosecond laser pulses interaction with gas/air-plasma has been studied for many years and several models have been proposed to interpret the experimental phenomena [1,2, and references within], including photocurrent model [3–5] and four-wave mixing [6,7]. According to the photocurrent model, when the electrons are released from the gas atoms by the laser field ionization, they absorb photon energy and are accelerated to produce transverse current which oscillates and emits THz wave pulse [3]. When the single-color laser pulse has the same amplitude as the two-color laser pulses, the ionization of atoms induced by them are nearly the same [4]. But the electron can be accelerated to a much larger velocity by two-color lasers than that by single-color laser. This is because the two-color laser pulses can break the symmetry of pulse field, so this process can produce

large net transverse photocurrent. And the THz generation from the two-color scheme is decided by the photocurrent directly, which means that the power law of THz yield as the function of the pump laser intensity is related to the production of photocurrent [8]. However, there are several ionization processes that are used to describe the atoms optical field ionization induced by strong laser pulses with varied laser intensity. For typical laser plasma interaction, its intensity range is from  $10^{12}$  to  $10^{16}$  W/cm<sup>2</sup> [9], and this means that the ionization includes multiphoton ionization (MPI) and tunneling ionization (TI). MPI and TI are two limitation cases of the atom ionization process. Especially, there is an evolution between them [10]. When the laser intensity varies, the laser field varies which makes the ionization process different. So the photocurrent and its THz generation induced by two-color laser pulses are affected by the optical ionization evolution directly. In this paper, the effect of this evolution on photocurrent is studied numerically based on the optical ionization processes.

## 2 Optical field ionization and photocurrent from two-color laser pulses

For short laser pulse, electrons are released from atoms or ions mainly by optical field ionization but not avalanche ionization. In the regime of strong laser field, MPI and TI are two ionization processes, which are characterized by Keldysh parameter [11–13].

$$\gamma_k = 2.31 \times 10^6 \left[ \frac{U_{\text{ion}}/\text{eV}}{(\lambda/\mu\text{m})^2 \cdot I/(\text{W} \cdot \text{cm}^{-2})} \right], \quad (1)$$

where  $U_{\text{ion}}$  is the atom ionization potential energy,  $\lambda$  and  $I$  are the laser wavelength and intensity, respectively. Equation (1) shows that  $\gamma_k$  is an intensity-dependent

parameter. For different  $\gamma_k$  of laser atom interaction, field ionization is dominated by MPI when  $\gamma_k \gg 1$  and TI when  $\gamma_k \ll 1$ . So if the laser wavelength and the gas atom species are set, the ionization process will be dependent on the laser intensity.

In the MPI regime, an electron escapes from the atom when it absorbs more than one photon to get enough energy. The ionization probability induced by per pulse cycle is very small and the ionization rate is a function of intensity which is described by [11]

$$w_{\text{MPI}} = \frac{2\pi\omega}{(l-1)!} \left( \frac{I}{I_{\text{mp}}} \right)^l, \quad (2)$$

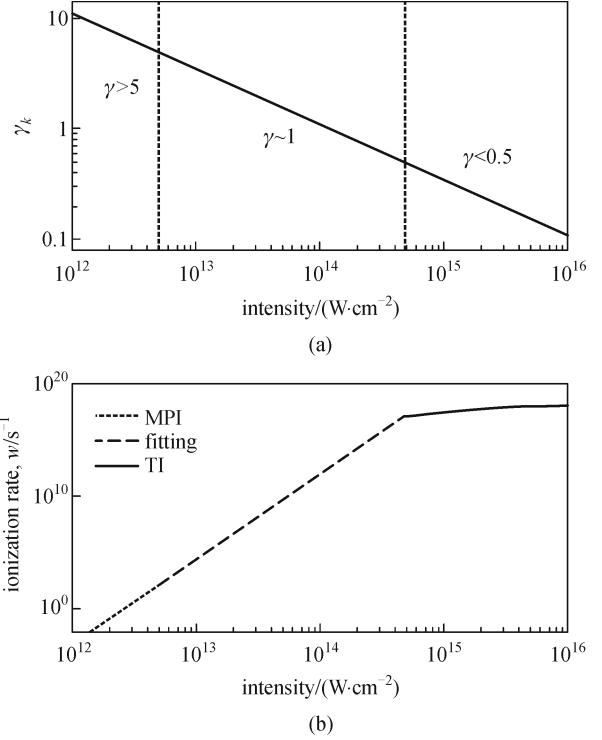
where  $\omega$  is the laser angle frequency,  $I$  is the laser intensity,  $l$  is the minimum number of photons needed for ionization (It is given by  $l = \text{Int}(U_{\text{ion}}/(\hbar\omega) + 1)$ ), and  $I_{\text{mp}} = \hbar\omega^2/\sigma_{\text{mp}}$  ( $\sigma_{\text{mp}}$  is cross section determined empirically to be equal to  $6.4 \times 10^{-18} \text{ cm}^2$ ). In the TI regime, according to Ammosov-Delone-Krainov (ADK) theory, the ionization rate is [11,12]

$$w_{\text{TI}} = 4\alpha\Omega_0 \left( \frac{U_{\text{ion}}}{U_{\text{H}}} \right)^{\frac{7}{4}} \left( \frac{I_{\text{H}}}{I} \right)^{-\frac{1}{4}} \exp \left[ -\frac{2}{3} \left( \frac{U_{\text{ion}}}{U_{\text{H}}} \right)^{\frac{2}{3}} \left( \frac{I_{\text{H}}}{I} \right)^{\frac{1}{2}} \right], \quad (3)$$

where  $\alpha$  is a fitting constant chosen to match the experimental measurement,  $\Omega_0 = 4.1 \times 10^{-16} \text{ s}^{-1}$  is the fundamental atomic frequency,  $U_{\text{H}}$  is the ionization energy of hydrogen atom, and  $I_{\text{H}} = 3.6 \times 10^{-16} \text{ W/cm}^2$ .

MPI ( $\gamma_k \gg 1$ ) and TI ( $\gamma_k \ll 1$ ) are two ionization limitation processes for different laser intensity. However, in the regime of  $\gamma_k \sim 1$  (with intensity  $10^{13} - 10^{14} \text{ W/cm}^2$ ), the ionization is neither purely MPI nor TI. These two processes exist simultaneously [14]. Although MPI is described well by the minimum-order perturbation theory, it is no longer convenient when the laser field becomes high enough. Then the ionization probability per cycle is still small but TI occurs during per cycle and is prominent [14]. Generally, an analytical fit with experiment data is used to calculate the total ionization rate induced by MPI and TI. Here a formula proposed by Kasparian et al. [15] is used to fit the ionization rate in the regime of  $\gamma_k \sim 1$ , which is  $w = R_{\text{T}}(I/I_{\text{R}})^{\alpha}$  with experimental value  $R_{\text{T}}$ ,  $I_{\text{R}}$  and  $\alpha$  who are dependent on the atom species. ( $\alpha = 7.5$  for nitrogen and  $\alpha = 6.5$  for oxygen). For the typical near infrared amplifier laser pulse with wavelength  $\lambda = 800 \text{ nm}$ , the intensity in the focus spot can reach up to  $10^{16} \text{ W/cm}^2$ , and its Keldysh parameter (Fig. 1(a)) and induced ionization rate (Fig. 1(b)) are shown in Fig. 1. Here the gas atom is set air atom. The principal constituents of air are nitrogen and oxygen, so the total  $\gamma_k$  and ionization rate  $w$  are written approximately by  $\gamma_k = 0.8\gamma_{\text{kN}_2} + 0.2\gamma_{\text{kO}_2}$  and  $w = 0.8w_{\text{N}_2} + 0.2w_{\text{O}_2}$  [13]. In the normal atmosphere nitrogen and oxygen have the proportion 4:1, so this

approximate is reasonable. From Fig. 1, it is found that in the low intensity, the ionization rate increases exponentially and then increase slowly in the TI regime. Between MPI and TI, the ionization rate is still an exponentially function of the laser intensity.



**Fig. 1** Relationship between Keldysh parameter  $\gamma_k$  (a), ionization rate  $w$  (b) and laser intensity

The density of electron after ionization is decided by the ionization rate and the time of ionization happened, which is given by

$$\frac{dn_e}{dt} = \sum_i w_i n_i, \quad (4)$$

where  $n_e$  is the electron density,  $w$  is the total ionization rate,  $n$  is the density of atom, and  $i$  is the different charges states. For the femtosecond laser pulse, the duration is several tens of femtoseconds, which is much shorter than the collision of particles. Therefore, here the collision of particles is neglected [4]. The released electrons are accelerated in the laser field by the Lorentz force and are calculated based on the classical mechanics [3]. When the laser intensity is limited in non-relativistic regime, the magnetic field is also neglected. So the velocity of electron at  $t$  in the linear polarized laser pulses is given by

$$v(t) = \int_{t_0}^t \frac{eE_l(t')}{m} dt', \quad (5)$$

where  $t_0$  is the time that electron is released from the atoms

or ions,  $e$  and  $m$  are the electron charge and mass, respectively.

Therefore, the transverse photocurrents at time  $T$  produced by laser pulse interaction with air can be calculated by

$$J_{\perp}(T) = \int_0^T e v d n_e = \frac{e^2}{m} \int_0^T \left[ \int_0^t E_i(t') dt' \sum_i w_i n_i \right] dt, \quad (6)$$

where  $\perp$  stands for the transverse. This means that only electron movement along the laser polarization contributes to the production of photocurrent. So in the non-relativistic intensity, when the laser pulses are set, the photocurrents produced by laser pulses interaction with air can be calculated from Eqs. (2) to (6). It has been found that the single color laser pulse cannot generate net large current because the laser field is symmetry and the electron cannot get the large velocity [3,4]. When the two-color scheme is used, the field symmetry is broken which makes the electron accelerated to large velocity in the laser field. Then this method can produce large net photocurrent. From the simulations, it is also found that this photocurrent can generate strong, broadband THz wave [4,16].

Here the photocurrents produced by two-color laser pulses interaction with air atoms are calculated. The pulses is given by

$$E_l = E_{\omega} \exp(-t^2/\tau^2) \cos(\omega t + \phi_1) + E_{2\omega} \exp(-t^2/\tau^2) \cos(2\omega t + \phi_2),$$

where  $\tau$  is the pulse duration,  $E_{\omega}$  and  $E_{2\omega}$  are the amplitudes of two pulses,  $\omega$  is the angle frequency,  $\phi_1$  and  $\phi_2$  are the phases of two pulses. In the simulations, the pulse duration is set  $\tau = 50$  fs, and the phases  $\phi_1 = \phi_2 = 0$ . For the MPI, the ionization rates caused by the fundamental pulse and second harmonic (SH) pulse is calculated separately, and then the free electrons are accelerated in the whole laser field. For the TI, the ionization rate is calculated by the total laser intensity, which includes fundamental pulse and SH pulse. Figure 2 is the relation of the photocurrent and the laser intensity. Here the current is normalized, so the density of air does not change the result. The ratio of  $E_{\omega}/E_{2\omega}$  in the simulations is set 19:1. The SH generation conversion rate is up to 6% in the two-color scheme, but there is a best conversion rate for the two-color scheme [1]. When the ratio of  $E_{\omega}/E_{2\omega}$  is changed, the two-color laser pulses shape is changed, which makes the net current different [4]. It shows that when the intensity is below  $10^{14}$  W/cm<sup>2</sup>, the currents increases exponentially with the intensity increasing, then it reaches nearly a constant. This is because in the lower intensity, both the electron density and velocity are increasing with the laser intensity. When the laser is becoming stronger, the gas atoms are ionized at all. Then

the increasing of the current is mainly induced by the increasing of the velocity. When the laser is stronger enough, the electrons are accelerated to a maximum value in the laser cycle. After that, even the laser intensity increase, the velocity of electron reaches a saturation which means saturation for photocurrent production. So when the power law of THz generation from the two-color scheme is investigated, the photocurrent production law should be considered. When the focus condition is changed, the laser intensity in air/gas plasma will be changed, and this makes the THz generation from it changed which has been detected in experiment [17]. As Fig. 2 shows, even the intensity changes a little in the range of  $10^{14}$  W/cm<sup>2</sup>, the current production changes greatly. So this relation affects THz generation power law directly.

When the phase difference between fundamental pulse and SH pulse changes, the total laser pulse shape is changed. Although the ionization rate is not affected, the acceleration of electrons is affected. So the photocurrent is also a function of the phase difference. Fixed the phase of fundamental pulse  $\phi_1$ , the photocurrent as a function of the SH phase  $\phi_2$  is calculated and plotted in Fig. 3. As it shows, the photocurrent is a periodic function of  $\phi_2$  at any regime of laser intensity. This is because with different intensity, the total laser pulses are similar even the magnitudes are different.

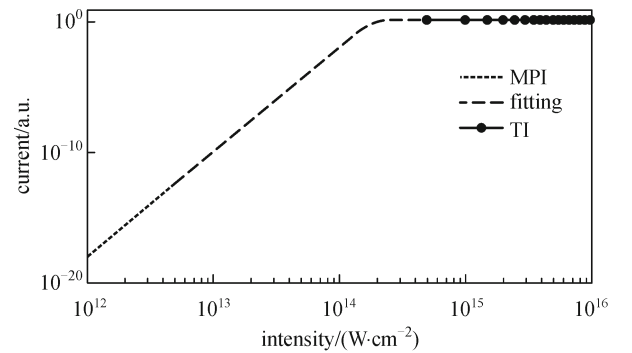
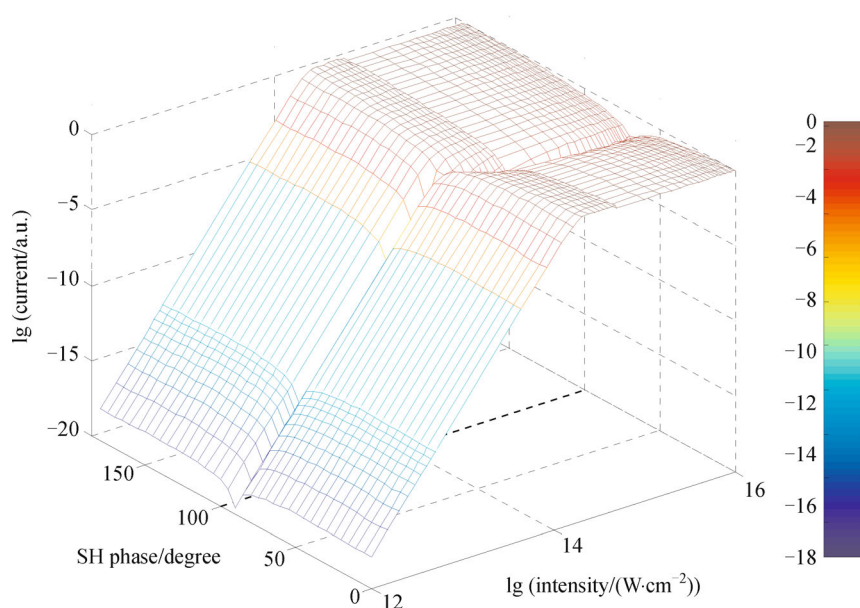


Fig. 2 Relationship between photocurrent and laser intensity

### 3 Conclusions

In conclusion, the ionization rate and the photocurrent of two-color scheme femtosecond laser pulses interaction with air are investigated. For typical laser plasma interaction process, the laser intensity is from  $10^{12}$  to  $10^{16}$  W/cm<sup>2</sup>, so the ionization is mainly MPI in the low regime and TI in the high regime. The gap between them is described by an empirical formula. Based on these ionization theories, the photocurrent produced from two-color femtosecond laser pulses interaction with air is calculated. It is found the photocurrent increases exponentially and then reaches a constant, which is because the



**Fig. 3** Relationship between laser intensity, second harmonic (SH) phase and photocurrent

electrons get saturation velocity in the laser cycle. This will be helpful to understand the power law of THz generation from the two-color laser scheme.

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## References

1. Thomson M, Kreß M, Löffler T, Roskos H. Broadband THz emission from gas plasmas induced by femtosecond optical: from fundamentals to applications. *Laser & Photonics Reviews*, 2007, 1(4): 349–368
2. Dai J, Clough B, Ho I, Lu X, Liu J, Zhang X C. Recent progresses in terahertz wave air photonics. *IEEE Transactions on Terahertz Science and Technology*, 2011, 1(1): 274–281
3. Du H. Terahertz wave generation and detection based upon ultrafast laser technology. *Chinese Journal of Quantum Electronics*, 2013, 30(3): 257–267
4. Kim K Y, Glowina J H, Taylor A J, Rodriguez G. Terahertz emission from ultrafast ionizing air in symmetry-broken laser fields. *Optics Express*, 2007, 15(8): 4577–4584
5. Du H, Chen M, Sheng Z, Zhang J. Numerical studies on terahertz radiation generated from two-color laser pulse interaction with gas targets. *Laser and Particle Beams*, 2011, 29(04): 447–452
6. Du H, Yang N. Effect of gas species on THz generation from two-color lasers. *Chinese Optics Letters*, 2013, 11(6): 063202
7. Xu R, Bai Y, Song L, Li N, Peng P, Liu P. Terahertz emission by balanced nonlinear effects in air plasma. *Chinese Optics Letters*, 2013, 11(4): 123002
8. Cook D J, Hochstrasser R M. Intense terahertz pulses by four-wave rectification in air. *Optics Letters*, 2000, 25(16): 1210–1212
9. Xie X, Dai J, Zhang X C. Coherent control of THz wave generation in ambient air. *Physical Review Letters*, 2006, 96(7): 075005
10. Li M, Li W, Shi Y, Lu P, Pan H, Zeng H. Verification of the physical mechanism of THz generation by dual-color ultrashort laser pulses. *Applied Physics Letters*, 2012, 101(16): 161104
11. Couairon A, Mysyrowicz A. Femtosecond filamentation in transparent. *Physics Reports*, 2007, 441(2–4): 47–189
12. Mevel E, Breger P, Trainham R, Petite G, Agostini P, Migus A, Chambaret J P, Antonetti A. Atoms in strong optical fields: Evolution from multiphoton to tunnel ionization. *Physical Review Letters*, 1993, 70(4): 406–409
13. Sprangle P, Peñano J R, Hafizi B. Propagation of intense short laser pulses in the atmosphere. *Physical Review E: Statistical, Nonlinear, and Soft Matter Physics*, 2002, 66(4): 046418
14. Keldysh L. Ionization in the field of a strong electromagnetic wave. *Journal of Experimental and Theoretical Physics*, 1965, 20: 1307–1314
15. Kasparian J, Sauerbrey R, Chin S. The critical laser intensity of self-guided light filaments in air. *Applied Physics. B, Lasers and Optics*, 2000, 71(6): 877–879
16. Dai H, Liu J. Terahertz emission dependence on the fundamental optical intensity in generating terahertz waves from two-color laser induced gas plasma. *Chinese Physics Letters*, 2011, 28(10): 104201
17. Blanchard F, Sharma G, Ropagnon X, Razzari L, Morandotti R, Ozaki T. Improved terahertz two-color plasma sources pumped by high intensity laser beam. *Optics Express*, 2009, 17(8): 6044–6052



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