The Role of Plasma Kinetic Processes during High Intense THz Pulses Generation

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Keywords: Two-color Laser Fields, Terahertz Generation from Plasma, Plasma Waves, Plasma Kinetics, Electron Velocity Distribution Function.

Abstract: This article is devoted to the theoretical study of THz radiation emission from plasma taking into account the kinetic properties of nonequilibrium electron velocity distribution function (EVDF) formed in the process of tunnel ionization by the two-color laser femtosecond pulse. The dispersive equation for longitudinal oscillations was solved within the framework of elementary and kinetic models of plasma volume. It was shown that the accounting of plasma kinetics can lead to essential variation of the spectral characteristics of generated pulse thus leading to a change in signal duration.

1 INTRODUCTION

The continuing over the years interest in sources of terahertz radiation is caused by manifold of their applications in various fields, such as spectroscopy, material science, biology, medicine, security systems and so on (Tonouchi, 2007; Nagai et al, 2006; Liu et al, 2010; Fischer et al, 2002; Kampfrath et al, 2013). Two-color schemes providing strong THz pulses with a very broad spectrum are the most popular among the plasma methods of THz generation (Thomson et al, 2010; Clerici et al, 2013; Oh et al, 2014; Andreeva et al, 2016). In this article we provide the generalization of the well-known transient photocurrent model of THz generation (Kim et al, 2007; Gildenburg and Vvedenskii, 2007; Silaev and Vvedenskii et al, 2009) by considering the kinetic approach for the electron ensemble relaxation during the THz waves emission. According to (Kim et al, 2007; Silaev and Vvedenskii et al, 2009) THz signal occurs in a region of the focal waist of two-color or ultrashort single-color laser field (it could be Ti-Sa or some other mid-IR laser source) caused by the residual electric photocurrent and polarization arising from the angular asymmetry of the photoionization process.

2 THz PULSE FORMATION IN PLASMA IN FRAMES OF ELEMENTARY MODEL

Let us start from the considering the emission from plasma within the elementary model. To produce plasma in our simulations we took two-color Ti-Sa laser pulse with the sine-squared envelope consisting of fundamental and second harmonics (400 + 800 nm) and gas xenon. The intensity of fundamental harmonic was chosen to be about $3.3 \cdot 10^{13}$ W/cm^2 (for the 2nd harmonic this value was four times less), pulse duration is about 100 fs, phase shift between the harmonics is $\pi/2$. The ionization process was considered in frames of tunnel ionization model (Delone and Krainov, 1998; Tong and Lin, 2005) (authors believe that this model is valid for xenon atom as well due to the proximity of xenon and hydrogen ionization potentials). The rate of ionization

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per unit of time in dependence of electric field strength $w(E)$ can be written in a form :

$$w(|E(t)|) = 4\omega_a(E_a/E)\exp(-2E_a/3|E(t)| - 12|E(t)|/E_a). \quad (1)$$

Here $E_a = 5.14 \cdot 10^9$ V/cm and $\omega_a = 4.13 \cdot 10^{16}\, s^{-1}$ are the atomic units of electric field and frequency, respectively. The temporal dynamics of the ionization process for the given two-color laser pulse is represented in Fig.1 for the xenon at atmospheric pressure. Thus, by the end of the laser pulse ionization is about $2.7 \cdot 10^{-4}$ which leads to the electron concentration about $N_e \cong 7 \cdot 10^{15}$ cm^{-3}.

The asymmetry of the ionization process in the two-color pulse leads to the formation of longitudinal plasma oscillations. To obtain the frequency domain of these oscillations one should solve the dispersive equation for longitudinal plasma waves:

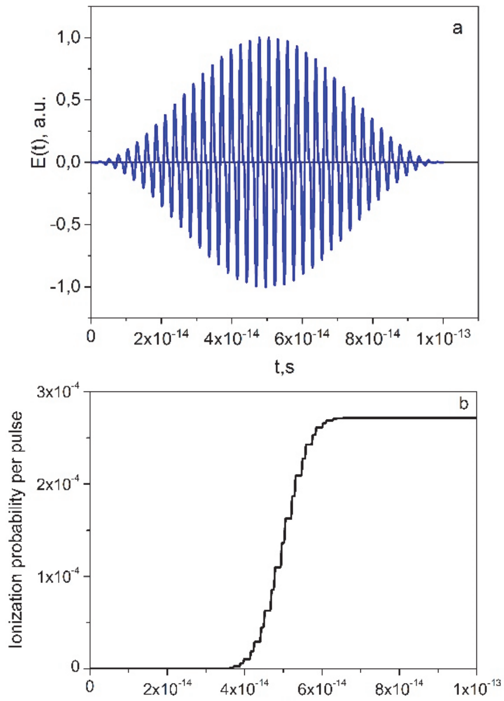

Figure 1: Time dependence of the ionization probability in xenon (b) under the action of the two-color Ti-Sa laser pulse (a). The parameters of laser pulse are listed in the text.

$$0 = \frac{\omega^2}{c^2}\varepsilon(\omega), \quad (2)$$

where $\varepsilon(\omega) = 1 + i\frac{4\pi\sigma(\omega)}{\omega}$ is the plasma permittivity at frequency ω and $\sigma(\omega)$ is the plasma conductivity. It should be mentioned that equation (2) is written for the longitudinal waves in free space. Considering the geometry of plasma volume where oscillations occur will lead to the following disperse equations (Landau and Lifshitz,1981):

$$0 = \frac{\omega^2}{c^2}(\varepsilon(\omega) + 1) \quad (3)$$

in the case of cylindrical plasma formation and

$$0 = \frac{\omega^2}{c^2}(\varepsilon(\omega) + 2) \quad (4)$$

for the spherical one.

In the frames of elementary model of plasma all electrons have equal velocities and $\sigma(\omega) = e^2N_e/m(\nu_{tr} - i\omega)$, where ν_{tr} denotes the transport collisional frequency of electrons in the focal volume which is assumed to have constant value and N_e is electron density in plasma. For typical laser plasma parameters the longitudinal plasma oscillations occur in the THz frequency band and, hence, can produce the seed THz pulse. By considering the cylindrical shape of plasma (this seems to be closer to the real shape of laser focal waist) and resolving equation (3) with respect to ω, one derives

$$\omega = \sqrt{\omega_p^2/2 - \nu_{tr}^2/4} - i\frac{\nu_{tr}}{2}. \quad (5)$$

Here $\omega_p = \sqrt{\frac{4\pi e^2 N_e}{m}}$ is the plasma (Langmuir) frequency which is about $5 \cdot 10^{12} s^{-1}$ for the above given electron density and belongs to THz frequency range. In order to produce signal of several periods of oscillations, attenuation in plasma should be less than the Langmuir frequency, thus in the first order with respect to the parameter $\frac{\nu_{tr}}{\omega_p} < 1$ one obtains from (5):

$$\omega \approx \frac{\omega_p}{\sqrt{2}} - i\frac{\nu_{tr}}{2}. \quad (6)$$

Real parts of the expressions (5) and (6) represent the frequency of electronic gas oscillations and it can be observed that taking into account the geometry of plasma volume leads to the shifting of this frequency while the imaginary part stands for the collisional attenuation of plasma wave. Evolution of plasma oscillations after the two-color laser pulse action can be described by plasma polarization:

$$P_s(z,t) = P_0 e^{-z^2/a^2} cos\left(\sqrt{\omega_p^2/2 - \nu_{tr}^2/4\, t}\right) e^{-\nu_{tr}/2t}. \quad (7)$$

Here P_0 is the value of residual polarization after the two-color pulse action, a is the size of focal waist of two-color laser pulse where the plasma waves are effectively excited (in our calculations we assume a is around 0.01 cm). Expression (7) was written under the assumption that in the region of pulse focal waist plasma oscillations take place with the same phase. Indeed, as the group velocity of the two-color pump pulse in plasma is close to the speed of light (here we neglect the walk-off between the fundamental and second pulse harmonics), one can consider the simultaneous excitation of plasma volume after the two-color pulse action. In case of proximity of plasma frequency and collisional frequency ν_{tr} the THz emission spectrum is wide consisting of different (both short and long) waves so the plasma can not be considered as a point-like source (especially for short waves). To obtain the emission of the seed pulse we are going to solve wave equation with the polarization (7) as the source:

$$\frac{\partial^2 E(z,t)}{\partial z^2} = \frac{1}{c^2}\frac{\partial^2 E(z,t)}{\partial t^2} + \frac{4\pi}{c^2}\frac{\partial^2 P(z,t)}{\partial t^2}. \quad (8)$$

By applying double Fourier transform to the equation (8): $E_{\omega,k} = \int_{-\infty}^{\infty} E(z,t)exp\,(i\omega t - ikz)dzdt$, $P_{\omega,k} = \int_{-\infty}^{\infty} P_s(z,t)exp\,(i\omega t - ikz)dzdt$ one obtains the following relation between the Fourier components of the polarization and the wave field:

$$E_{\omega,k}(k^2 - \omega^2/c^2) = \frac{4\pi}{c^2}\omega^2 P_{\omega,k}, \quad (9)$$

where

$$P_{\omega,k} = -iP_0\frac{a}{2}\sqrt{\pi}\exp\left(-\frac{a^2k^2}{4}\right)\times \left(1\Big/\left(\left(\omega - \sqrt{\omega_p^2 - \nu_{tr}^2/4}\right) + \frac{i\nu_{tr}}{2}\right) + \right. \left. + 1\Big/\left(\left(\omega + \sqrt{\omega_p^2 - \nu_{tr}^2/4}\right) + \frac{i\nu_{tr}}{2}\right)\right) \quad (10)$$

From (9) the spatial evolution of the plasma emission spectrum reads:

$$E_\omega(z) = \frac{1}{2\pi}\int_{-\infty}^{\infty} 4\pi\frac{\omega^2}{c^2}\frac{P_{\omega,k}}{k^2-\omega^2/c^2}exp(ikz)dk. \quad (11)$$

The solution $E_\omega(z)$ of equation (9) for $z = 0$ will represent the spectrum which is formed from the oscillations of polarization vector (7). Expression (11) can be resolved by means of residue theory. By choosing only the wave propagating in positive direction for the pulse spectrum we finally have:

$$E_\omega(z)|_{z=0} = P_0 a\pi^{\frac{3}{2}}\frac{\omega}{c}exp\left(-\frac{\omega^2 a^2}{4c^2}\right)\times \left(1\Big/\left(\left(\omega - \sqrt{\omega_p^2 - \nu_{tr}^2/4}\right) + \frac{i\nu_{tr}}{2}\right) + \right. \left. 1\Big/\left(\left(\omega + \sqrt{\omega_p^2 - \nu_{tr}^2/4}\right) + \frac{i\nu_{tr}}{2}\right)\right). \quad (12)$$

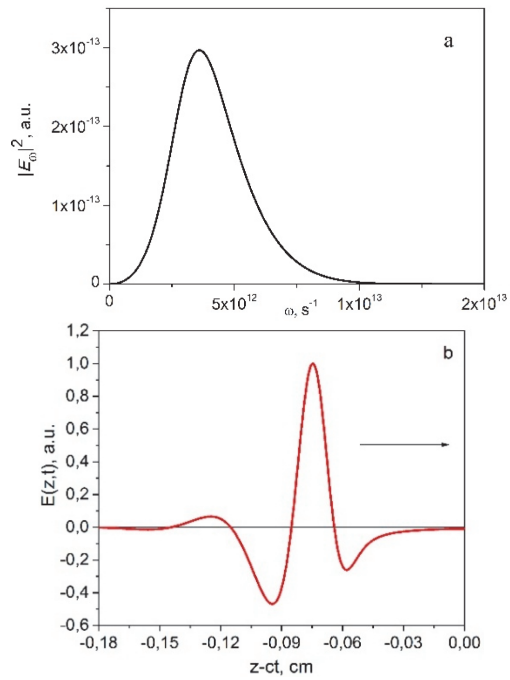

Figure 2: Spectrum (a) and spatial-temporal distribution (b) of emitted signal in plasma within the elementary model. Plasma electron concentration $N_e = 7\cdot 10^{15}$ cm^{-3}. Transport frequency in plasma $\nu_{tr} = 3.75\cdot 10^{12}$ s^{-1}. Arrow shows the direction of pulse propagation.

Typical spectrum of emission $|E_\omega|^2$ and spatial-temporal distribution of the produced pulse are shown in Figure 2 for the plasma frequency $\omega_p = 5\cdot 10^{12}$ s^{-1} (which corresponds to the electron concentration $N_e = 7\cdot 10^{15}$ cm^{-3}), $\nu_{tr} = 3.75\cdot 10^{12}$ s^{-1} (for estimates we selected gas xenon at atmospheric pressure and the velocity of electrons was chosen to be about 10^8 cm/s).

3 THz PULSE FORMATION UNDER THE INFLUENCE OF KINETIC PROPERTIES OF NONEQUILIBRIUM PLASMA

The main issue of this section is the study of influence of plasma kinetic properties on the THz pulse formation. Within the framework of the two-term expansion for Boltzmann kinetic equation the plasma conductivity can be expressed as (Ginzburg and Gurevich, 1960; Raizer, 1977):

$$\sigma(\omega) = \frac{{\omega_p}^2}{3}\int_0^{\infty}\frac{v^3(\nu_{tr}(v)+i\omega)}{\omega^2+\nu_{tr}^2(v)}\left(-\frac{\partial f}{\partial v}\right)dv, \quad (13)$$

where $f(v)$ stands for electron distribution function over absolute value of velocity normalized by $\int_0^\infty f(v)v^2 dv = 1/4\pi$. Substituting (13) in dispersive equation (3) for the cylindrical plasma geometry one obtains the following integral equation for longitudinal plasma oscillations:

$$\omega = -\frac{1}{2} i \frac{4\pi}{3} \omega_p^2 \int_0^\infty \frac{v^3(\nu_{tr}(v)+i\omega)}{\omega^2+\nu_{tr}^2(v)} \left(-\frac{\partial f}{\partial v}\right) dv. \qquad (14)$$

Solution of the equation (14) provides the spectrum of plasma waves for the given electron velocity distribution function (EVDF) and in general terms should be solved numerically. In case of $\omega_p > \langle \nu_{tr} \rangle$,where $\langle \quad \rangle$ means averaging over the distribution function, it is possible to write the analytical solution in the first order over parameter $\langle \nu_{tr} \rangle / \omega$:

$$\omega \cong \frac{\omega_p}{\sqrt{2}} - \frac{i}{2} \cdot (< \nu_{tr}(v) > + \\ + \frac{1}{3} < v \frac{\partial \nu_{tr}(v)}{\partial v} >). \qquad (15)$$

It can be seen that accounting of plasma kinetics leads to additional term in the imaginary part of ω compared to (6) which could be both positive or negative depending on the sign of the derivative function $\frac{\partial \nu_{tr}(v)}{\partial v}$ and the expression for the EVDF. Thus, taking into account both the velocity dependence of transport scattering cross section and distribution of electrons enables to describe the spectrum of emitted radiation from plasma more correctly.

Figure (3) demonstrates the electron velocity distribution functions formed along the polarization axis during the tunnel ionization of xenon by two-color femtosecond laser pulses with fundamental wavelengths of 800 and 3900 nm. The duration of 3900 nm pulse and the intensities of fundamental and second harmonics were chosen to be the same as for the 800 nm pulse which was considered in the Section I. Both obtained distributions are highly nonequilibrium and have sharp angular distribution along the direction of the electric field of the pulse. Below we will call them EVDF$_1$ for the distribution formed by the (400+800) nm laser pulse and EVDF$_2$ for the distribution formed by the (1950+3900) nm laser pulse. According to the plasma kinetic theory, elastic collisions lead to the isotropization of angular distribution of electrons, while the domination of electron-electron collisions determines fast maxwellization of EVDF. Duration of the maxwellization process can be estimated as $\tau_M \sim \nu_{ee}^{-1}$, where ν_{ee} is the frequency of electron - electron collisions. By estimating electron-electron collisional cross section as $\sigma_{ee} \approx (\pi e^4/\varepsilon_e^2) \times L$ (here ε_e is the electron energy and $L \approx 10$ is the Coulomb logarithm) for $\varepsilon_{e1} \sim 0.5$ eV which corresponds to maximum of EVDF$_1$ (see fig.4) and $\varepsilon_{e2} \sim 12.4$ eV for the EVDF$_2$, one obtains $\tau_{M1} \sim 1.4 \times 10^{-11}$ s^{-1} and $\tau_{M2} \sim 1.6 \times 10^{-9}$ s^{-1}. Here N_e was chosen to be 7×10^{15}cm^{-3} (obtained by the formula (1) ionization degree is approximately the same for both cases). The above estimates lead to the fact that for the EVDF$_1$ fast maxwellization will take place, while for the EVDF$_2$ elastic collisions will redistribute electrons rather quickly (within the time interval of several collisions) leading to the isotropic distribution in velocity space. For the transport collisional frequencies we derive $\nu_{tr1} \approx 10^{11}$ s^{-1} and $\nu_{tr2} \approx 6.6 \times 10^{12}$ s^{-1}.

To estimate the temperature T of the maxwellized EVDF$_1$ one can write the following relation

$$\frac{1}{2}\frac{m(\Delta v_{||})^2}{2} + \frac{m(v_0)^2}{2} + 2 \cdot \frac{1}{2}\frac{m(\Delta v_\perp)^2}{2} = \frac{3}{2} T. \qquad (16)$$

which has the sense of energy conservation law during the isotropization process in three-dimensional velocity space. Here v_0, $\Delta v_{||}$ are taken from Fig. 3, $\Delta v_\perp$ stands for the transverse distribution of photoelectrons formed in the tunnel ionization, which is usually assumed to be small compared with the electron velocities along the pulse polarizations (Delone and Krainov, 1998). In Fig. 4 the obtained isotropic distributions are plotted with the function $v \frac{\partial \nu_{tr}(v)}{\partial v}$ for xenon to analyse the contribution of kinetic effects to the imaginary part of solution (15),

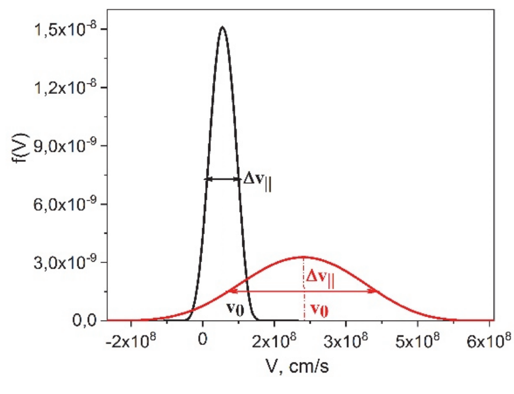

Figure 3: One-dimensional electron velocity distribution functions (EVDF) along the polarization of two-color laser field with fundamental wavelengths 800 nm (black curve) and 3900 nm (red curve). Here v_0, $\Delta v_{||}$ are the mean velocity and velocity dispersion correspondingly.$v_0 = 4.2 \cdot \frac{10^7 \text{см}}{c}$, $\Delta v_{||} = 5.3 \cdot 10^7$ cm/s for 800 nm; $v_0 = 2.09 \cdot 10^8$ cm/s, $\Delta v_{||} = 2.46 \cdot 10^8$ cm/s for 3900 nm. Distribution functions are normalized to unity.

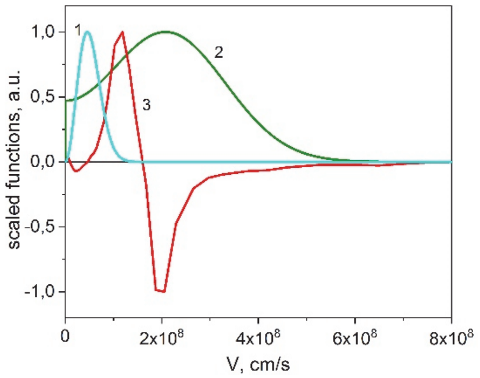

Figure 4: EVDF$_1$ after the Maxwellization process (1), EVDF$_2$ after the isotropization process (2) and $v\frac{\partial v_{tr}(v)}{\partial v}$ (3). All the functions are scaled (divided by maximum values). EEDF$_1$ and EEDF$_2$ represent the velocity distribution in three-dimensional velocity space and normalized by condition $\int f(v)v^2dv = 1/4\pi$.

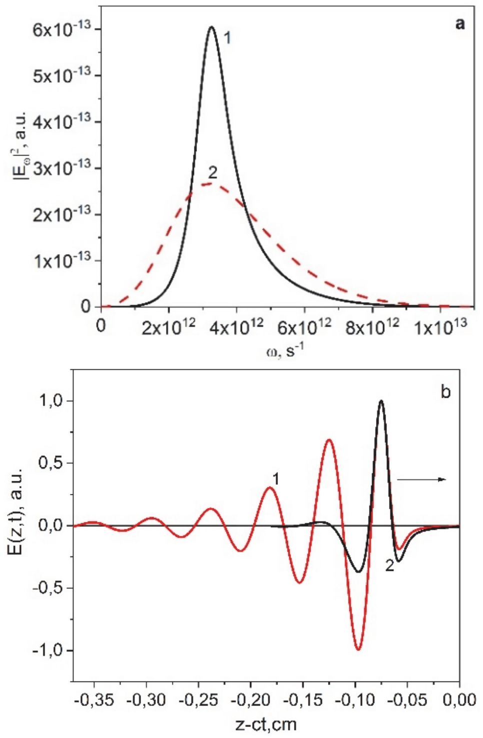

Figure 5: Spectra of emission (a) and spatial-temporal distributions of electric field strength (b) for plasma characterized by the distribution functions EVDF$_1$ (1) and EVDF$_2$ (2).

which determines the collisional damping of plasma waves. The value of transport scattering cross section for xenon atom was taken from (Hayashi, 1983). As for distribution (2) in Fig. 4 it comes from the distribution formed by the (1950+3900) nm laser pulse replotted for the velocity module and renormalized in three-dimensional velocity space. One can see, for example, that for the distribution (2) the negative value of function $v\frac{\partial v_{tr}(v)}{\partial v}$ can decrease the value of damping coefficient in (15), thereby leading to the formation of longer THz pulses with narrower spectrum.

In case of our consideration plasma frequency is close to the transport collisional frequency, so we apply numerical methods to the solution of (14). Equation (14) was solved by the iterative method, and for obtained distribution functions we have the following results:

$\omega \approx 3.3 \cdot 10^{12} - 4.3 \cdot 10^{11} i$ s^{-1}	for the EVDF$_1$
$\omega \approx 2.5 \cdot 10^{12} - 1.9 \cdot 10^{12} i$ s^{-1}	for the EVDF$_2$.

The spectra of emission $|E_\omega|^2$ and spatial-temporal distributions of electric field strength $E(z,t)$ for both distribution functions are shown in Fig.5.

4 CONCLUSIONS

This article considers the THz pulse generation through the longitudinal plasma oscillations arising from gas ionization by two-color femtosecond laser pulse. The obtained analytical and numerical results demonstrate that due to the strong velocity dependence of transport scattering cross section of gas atoms various velocity distributions of plasma electrons can provide sufficiently different spectra of emission, which can lead to generation of THz pulses with different spectral and spatial widths. The performed research will play important role for the new approach to obtain high intense THz pulses in nonequilibrium plasma of noble gases proposed and developed by the authors [Bogatskaya and Popov, 2013; Bogatskaya et al, 2014; Bogatskaya and Popov, 2018]. The approach consists in additional amplification of seed THz pulse which arises under the action of two-color (mid)IR laser in photoionized plasma channel formed by femtosecond eximer KrF laser pulse.

ACKNOWLEDGEMENTS

This work was supported by the Russian Science Foundation (project no. 18-72- 00125).

REFERENCES

Andreeva, V., A., Kosareva, O., G., Panov, N., A., Shipilo, D., E., Solyankin, P., M., Esaulkov, M., N., González, P., Shkurinov, A., P., Makarov, V., A., Bergé, L., Chin, S. L., 2016. Phys. Rev. Lett., 116, 063902.

Bogatskaya, A., V., Popov, A., M., 2013. JETP Lett. 97, 388

Bogatskaya, A., V., Popov, A., M., Smetanin, I., V., 2014. J. Rus. Las. Res., 35, 437.

Bogatskaya, A., V., Popov, A., M., 2018. Las. Phys., 28, 115301

Clerici, M., et al 2013. Phys. Rev. Lett., 110, 253901.

Delone, N., B., Krainov, V., P., 1998. Phys. Usp., 41, 469.

Fischer, B., M., Walther. M., Jepsen.,P., U., 2002. Phys. Med. Biol., 47, 3807.

Gildenburg, V., B., Vvedenskii N., V., 2007. Phys. Rev. Lett. 98, 245002.

Ginzburg, V., L., Gurevich, A., V., 1960. Usp. Fiz. Nauk, 70, 201.

Hayashi, M., 1983. J. Phys. D., 16, 581.

Kampfrath, T., Tanaka, K., Nelson, K., A., 2013. Nat. Photonics, 7, 680.

Kim, K., Y., Glownia, J., H., Taylor, A., Rodriguez, J., G., 2007. Opt. Express, 15, 4577.

Landau, L., D., Lifshitz, L., M., 1981. *Quantum Mechanics: Non-Relativistic Theory*, Butterworth Heinemann, 3rd Edition.

Liu, J., Dai J, Chin, S., L., Zhang, X., C., 2010. Nat. Photon., 4, 627.

Nagai, N., Sumitomo, M., Imaizumi, M., Fukasawa, R., 2006. Semicond. Sci. Technol., 21, 201.

Oh, T., I., Yoo, Y., J., You Y., S., Kim, K., Y., 2014. Appl. Phys. Lett., 105, 041103.

Raizer, Yu., P., 1974. *Laser-Induced Discharge Phenomena*, Consultants Bureau, New York, London.

Silaev, A., A., Vvedenskii, N., V., 2009. Phys. Rev. Lett., 102, 115005.

Thomson, M., D., Blank, V., Roskos, H., G., 2010. Opt. Express, 18, 23173.

Tong, X., M., Lin, C., D., 2005. J. Phys. B, 38, 2593.

Tonouchi, M., 2007. Nat. Photon., 1, 97.