*Quantum Electronics* **46** (4) 338–341 (2016) 

DOI: 10.1070/QEL16055

# X-ray spectral diagnostics of laser harmonic generation in the interaction of relativistic femtosecond laser pulses with clusters

A.Ya. Faenov, E. Oks, E. Dalimier, I.Yu. Skobelev, S.A. Pikuz, T.A. Pikuz, I.A. Zhvaniya, Y. Fukuda, A. Andreev, J. Koga, H. Sakaki, H. Kotaki, A. Pirozhkov, Y. Hayashi, T. Kawachi, M. Kando, K. Kondo, A. Zhidkov, R. Kodama

***Abstract.*** **It is shown that the production of X-ray emission spectra in the interaction of high-intensity laser radiation with cluster targets may be affected by the bichromatic oscillating electric field arising from the generation of the second harmonic of laser radiation. A technique is proposed for diagnosing harmonic generation in laser–cluster interactions using the spectral line profiles of multiply charged helium ions. The efficiency of second harmonic generation at a laser intensity of $3\times10^{18}$ W cm$^{-2}$ is shown to amount to about 2%.**

***Keywords:*** *relativistic femtosecond laser pulses, clusters, harmonic generation, X-ray diagnostics.*

Despite the fact that X-ray spectral plasma diagnostics came into being a long time ago, even now it is successfully employed for determining the parameters of high-temperature plasmas – plasmas produced in different laboratory facilities as well as plasmas existing in astrophysical objects (see, for instance, Refs [1–12]). In this case, advantage is taken of the circumstance that the plasma emission spectrum depends on plasma parameters, and it is possible to select the parameter values whereby the simulated emission spectrum is consistent with the observed one.

The X-ray radiation spectra of a high-temperature plasma are formed primarily due to atomic processes occurring in it, like electron–ion collisions and the radiative or autoionisation decay of ion levels. Since the probabilities of these events depend mostly on the electron temperature and density, X-ray methods may be employed primarily for estimating the electron density and temperature of the plasma.

The existence of slowly varying (quasi-static) electric and magnetic fields in the plasma also exerts influence on its radiative characteristics and changes the profiles of radiated spectral lines due to Zeeman and Stark effects. Accordingly, measurements of spectral line shapes permit revealing the quasi-static electromagnetic fields existing in the plasma.

Furthermore, the plasma may be the site of rather high oscillating electromagnetic fields, which are due to external action on the plasma as well as due to the emergence of strong plasma instabilities and their corresponding plasma oscillations. For instance, the properties of a plasma produced by high-power ultrashort laser pulses arise from the existence of a strong anisotropy in electron velocity distribution. A consequence of this anisotropy is the emergence of strong electromagnetic oscillations caused by the development of Weibel instability. This leads to the generation of strong quasi-stationary magnetic fields with intensities depending on the intensity of laser radiation. The existence of the magnetic field may in turn result in the generation of Bernstein oscillations. Another example is provided by high-frequency electron Langmuir oscillations. Additional mechanisms may be of significance in experiments with clusters. In this case, strong oscillations may develop also at frequencies $\omega_p = 3\omega_{las}$ because of the final target size.

A characteristic spectroscopic manifestation of the presence of a monochromatic oscillating electric field in the plasma is the emergence of additional spectral lines (satellites) with frequencies $\omega_s = \omega_a \pm m\omega_{las}$, where $\omega_a$ is the atomic transition frequency; $m$ = 1, 3, 5,... for a parity-forbidden atomic transition; and $m$ = 2, 4,... for a parity-allowed transition. These satellites are termed laser or plasma satellites, depending on whether the oscillating field is an external laser field or plasma oscillations. When the atomic transition frequency lies in the X-ray range of the spectrum, the resultant satellites will not, as a rule, be isolated lines and will form the profile of the observed X-ray transitions.

X-ray plasma satellites were earlier observed in the interaction of moderate-intensity picosecond laser pulses with solid targets [13–14] and in the interaction of femtosecond laser pulses of higher intensity with cluster targets [15]. In this case, the theoretical treatment was restricted to the case of monochromatic electromagnetic fields.

However, for very high (relativistic) intensities of laser radiation, nonlinear processes occurring in the laser plasma should give rise to electromagnetic oscillations with frequencies which are multiples of the laser frequency, i.e. to the generation of harmonics of the heating laser radiation. Among

**A.Ya. Faenov, I.Yu. Skobelev, S.A. Pikuz, I.A. Zhvaniya** Joint Institute for High Temperatures, Russian Academy of Sciences, Bldg. 2, ul. Izhorskaya 13, 125412 Moscow, Russia; e-mail: spikuz@gmail.com;
**E. Oks** Physics Dept., Auburn University, Auburn, AL 36849, USA;
**E. Dalimier** LULI-UPMC Univ Paris 06: Sorbonne Universites; CNRS, Ecole Polytechnique, CEA: Universite Paris-Saclay – F-75252 Paris Cedex 05, France;
**T.A. Pikuz** Joint Institute for High Temperatures, Russian Academy of Sciences, Bldg. 2, ul. Izhorskaya 13, 125412 Moscow, Russia; present address: Institute for Academic Initiatives, Osaka University, Osaka, 565-0871, Japan;
**Y. Fukuda, J. Koga, H. Sakaki, H. Kotaki, A. Pirozhkov, Y. Hayashi, T. Kawachi, M. Kando, K. Kondo** Quantum Beam Science Directorate, Japan Atomic Energy Agency, Kyoto 619-0215, Japan;
**A. Andreev** Max Born Institute, Berlin 12489, Germany; ELI-ALPS, Szeged H-6720, Hungary;
**A. Zhidkov** PPC Osaka University and JST, CREST, Osaka 565-0871, Japan;
**R. Kodama** Institute for Academic Initiatives, Osaka University, Osaka, 565-0871, Japan; PPC Osaka University and JST, CREST, Osaka 565-0871, Japan

Received 24 February 2016
*Kvantovaya Elektronika* **46** (4) 338–341 (2016)
Translated by E.N. Ragozin

possible scenarios is the following one. A transverse laser wave $t_1$ excites a longitudinal Langmuir wave $l$ and an ion-acoustic wave $t_1 \to l + s$. Then, a transverse wave $t_2$ at the double laser frequency is produced due to one of the two processes: $l + l \to t_2$ or $l + t_1 \to t_2$. An important result of these nonlinear processes is that the plasma ions, which emit X-ray spectral lines, will experience a bichromatic field (the sum of two monochromatic waves with frequencies $\omega_{\rm las}$ and $2\omega_{\rm las}$).

The behaviour of a radiating ion in such a bichromatic field was first investigated by Oks et al. [16], who showed that the ion emission spectrum in this case exhibits a nontrivial dependence on the amplitude ratio of the first ($E_1$) and second ($E_2$) harmonics. Some or other plasma satellites may be prevalent in the emission spectrum, depending on precisely the ratio $E_1/E_2$. In a bichromatic field, the relative intensities of the satellites shifted from the centre by the laser frequency $\omega_{\rm las}$ and the double laser frequency $2\omega_{\rm las}$ are expressed by the sum of the products of Bessel functions, whose argument is proportional to the ratios $nE_1/\omega_{\rm las}$ and $nE_2/(2\omega_{\rm las})$, where $n$ is the principal quantum number of the energy level which gives rise to one or other spectral line. Since Bessel functions are oscillating functions, also oscillating are the relative intensities of the satellites at frequencies $\omega_{\rm las}$ and $2\omega_{\rm las}$. As a result, observed for some $E_2/E_1$ ratios is the suppression of plasma lines at the frequency $\omega_{\rm las}$. Moreover, for some ratios a situation is possible whereby the satellites at the first harmonic frequency are suppressed even when $E_1 > E_2$. In this case, the combined analysis of two spectral lines characterised by different values of $n$ permits an unambiguous determination of the ratio $E_2/E_1$ for the experimental conditions under considerations.

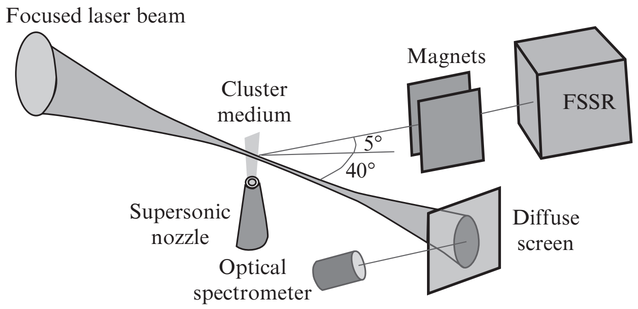


**Figure 1.** Experimental setup.

From the aforesaid it follows that a comparison of the observed spectrum with the simulated ones may permit us not only to detect the very fact of second harmonic generation but also to estimate its intensity relative to the fundamental intensity. In this work we employed the approach of Ref. [16] to interpret the results of experiments on the heating of argon clusters by femtosecond laser pulses with an intensity of up to $3\times10^{18}$ W cm$^{-2}$.

Our experiments were carried out in the Kansai Photon Science Institute of Japan Atomic Energy Agency (KPSI, JAEA, Japan). Use was made of two Ti:sapphire laser facilities with a radiation wavelength of 800 nm. In one case, the JLITE-X laser generated an output pulse with a duration of

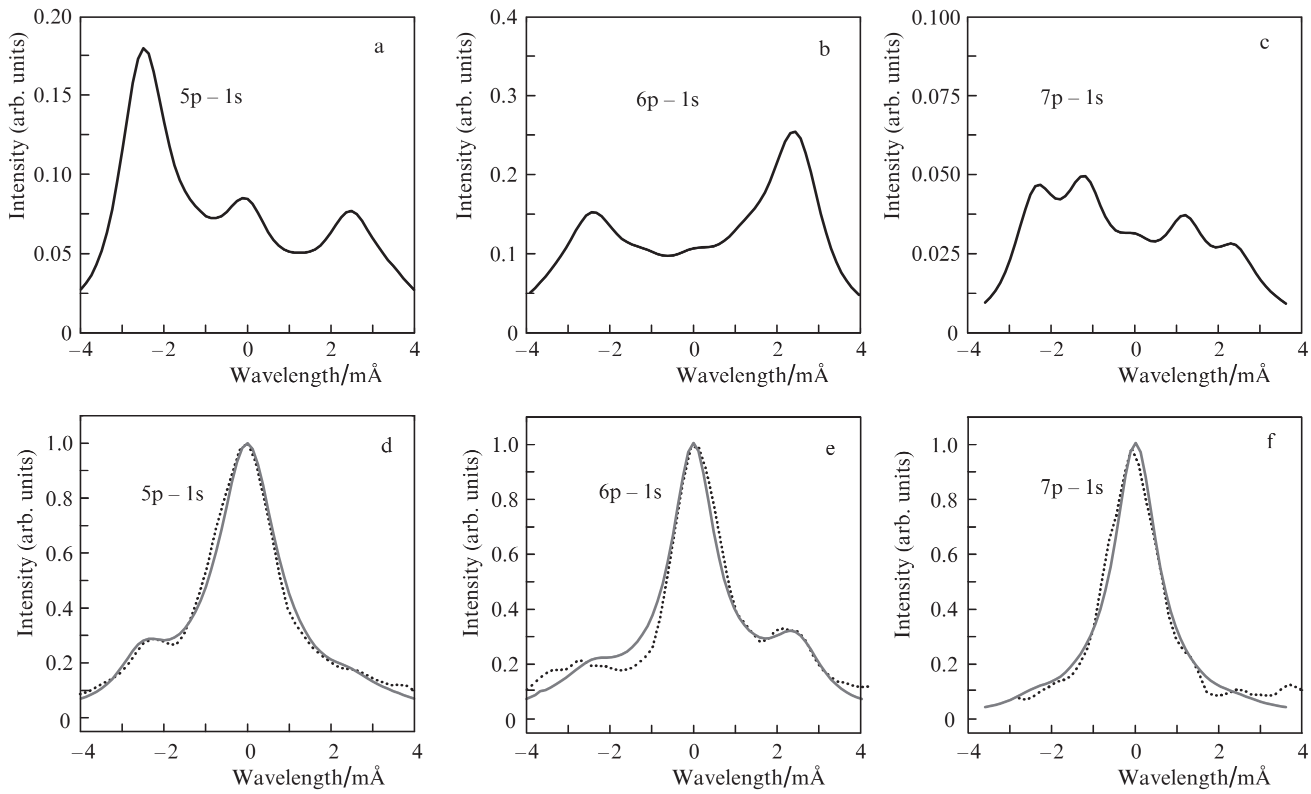


**Figure 2.** Comparison of the experimental and simulation ArXVII line spectra arising from the 5p–1s ($He_5$), 6p–1s ($He_6$) and 7p–1s ($He_7$) transitions. The experimental data were obtained on the J-KAREN facility for a laser intensity of $3\times10^{18}$ W cm$^{-2}$. The simulation spectra (a–c) were obtained for a relatively small plasma area with a bichromatic field $E_1\cos(\omega t) + E_2\cos(2\omega t)$ ($E_1$ = 14 GV cm$^{-1}$, $E_2$ = 7 GV cm$^{-1}$) [16]. Figures 2d–2f depict the sums of the simulation spectra [16] radiated from the relatively small area with the bichromatic field and the relatively large area without periodic fields, as well as experimental spectra (dotted curves).

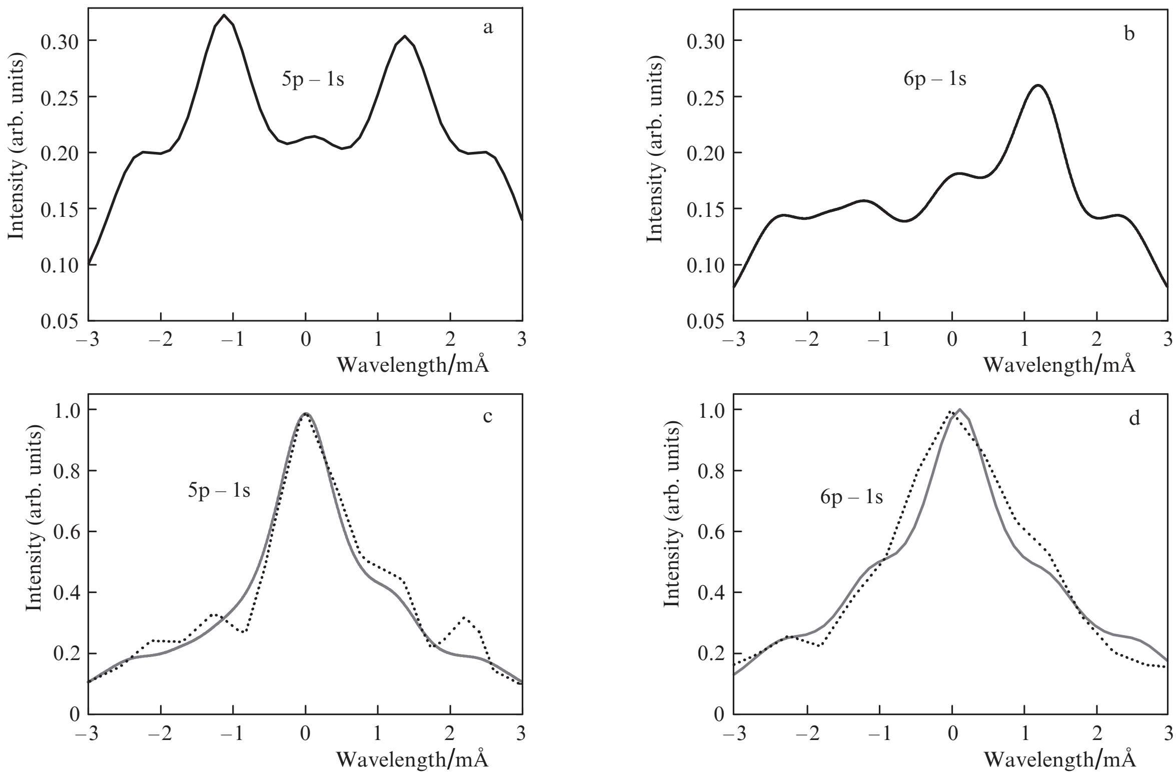


**Figure 3.** Comparison of the experimental and simulation ArXVII line spectra arising from the 5p – 1s ($He_5$) and 6p – 1s ($He_6$) transitions recorded for a laser intensity of $4\times10^{17}$ W cm$^{-2}$ and a pulse duration of 500 fs (the JLITE-X facility). The curves in Figs 3a and 3b represent the simulation spectra from the relatively small plasma area with the bichromatic field $E_1\cos(\omega t) + E_2\cos(2\omega t)$ ($E_1$ = 14 GV cm$^{-1}$, $E_2$ = 7 GV cm$^{-1}$). The solid curves in Figs 3c and 3d are the sums of the simulation spectra radiated from the small area with the monochromatic field and the relatively large area with no periodic fields present; the dotted curves stand for experimental spectra.

40 fs, an energy of 160 mJ, and a contrast ratio of $10^5$, which permitted conducting experiments at intensities up to $4\times10^{17}$ W cm$^{-2}$ [17]. In the other case, advantage was taken of the J-KAREN laser [18], which provided a far higher contrast ratio of $10^8$ – $10^{10}$ (on a picosecond time scale). This improvement of the contrast ratio was achieved by placing a saturable absorber between the master oscillator and the stretcher. An ultrafast Pockels cell provided improvement of the contrast ratio on a nanosecond scale. The output pulse of this facility, also 40 fs in duration, had an energy of 800 mJ, which produced an intensity of $3\times10^{18}$ W cm$^{-2}$ in a focal spot 30 μm in diameter [19]. In both cases, the laser beam was focused with an off-axis parabolic mirror into an argon cluster jet (Fig. 1).

Argon clusters were produced when the gas with a high initial pressure expanded in vacuum through a specially developed supersonic nozzle [20, 21], which consisted of three coaxial conical surfaces. Our experiments were mainly performed for an initial pressure of 6 MPa, when the average Ar cluster size was equal to about 1.5 μm in the laser focal spot area. The combination of a large cluster size and a high contrast ratio of laser pulses provided survival of the dense cluster core until the arrival of the main pulse, i.e. the laser prepulse did not disrupt the clusters completely.

The plasma emission spectra were recorded with a high spectral and spatial resolution using a focusing spectrometer with spatial resolution (FSSR) [22 – 24]. The spectrometer made use of spherically bent quartz crystals ($2d$ = 4.912 Å, radius of curvature $R$ = 150 mm). The spectrometer was tuned to a range of 3.05 – 3.5 Å, which hosted the spectral lines of He-like ArXVII (the resonance series 1snp – 1s$^2$ for $n$ = 3 – 7). The spectral resolving power $\lambda/\Delta\lambda$ was no worse than 3000.

The lineouts of the spectra obtained for a laser intensity of $3\times10^{18}$ W cm$^{-2}$ are exemplified in Fig. 2. The profiles of spectral lines $He_5$ and especially $He_6$ clearly exhibit peaks (plasma satellites) separated from the main lines by $2\omega_{las}[\lambda_n^2/(2\pi c)]$ in wavelength, while the plasma satellites shifted by the fundamental frequency are absent. If the radiation were collected from the plasma area where only the $t_2$ wave existed, the interpretation of experimental spectra would be quite evident. But this could be the case if only the above-critical plasma with a density $N_e$ ranging from $N_{cr}$ to $4N_{cr}$ were radiating, where $N_{cr}$ = $1.8\times10^{21}$ cm$^{-3}$ for the laser frequency $2.4\times10^{15}$ s$^{-1}$. For these densities, however, the Stark widths of the lines $He_5$, $He_6$, etc. would be much broader than the observed ones. Therefore, it must be admitted that the wave $t_1$ is also present, along with the wave $t_1$, in the area responsible for the emission of the spectra and that its intensity exceeds the intensity of the $t_2$ wave.

The theory of plasma satellites in a bichromatic field was recently proposed and elaborated in Ref. [16]. It suggests that the intensity of the plasma satellite shifted by the fundamental frequency may be far lower than that of the satellite shifted by the double fundamental frequency for some ratio of the amplitudes $E_1$ and $E_2$. Since this is experimentally observed for at least two transitions, $He_5$ and $He_6$, it is possible to find not merely the $E_1/E_2$ amplitude ratio, but the absolute values of each amplitude as well. For instance, to reach agreement between the experimental and simulation profiles in the case shown in Fig. 2, we have to assume that $E_1$ = 14 GV cm$^{-1}$ and

$E_2 = 7$ GV cm$^{-1}$. Since the amplitude of the electric field of the laser wave in vacuum for an intensity of $3 \times 10^{18}$ W cm$^{-2}$ is $E_0 = 48$ GV cm$^{-1}$, the efficiency of laser-to-second harmonic radiation conversion efficiency amounted to 2% in this experiment. This was also borne out by the particle-in-cell simulations performed in Ref. [16].

In the simulation of experimental profiles of spectral lines, account must be taken of the fact that the observed radiation is a superposition of the radiation emanating from the dense plasma area, which is the site of the bichromatic field with $t_1$ and $t_2$, and the radiation from a lower-density plasma area of greater size with no oscillating electric fields. The higher density area forms a relatively broad pedestal about every spectral line, while the lower density area radiates a substantially narrower component of higher intensity. These integral profile are also plotted in Fig. 2. The best agreement between the experimental and simulation profiles was reached when the electron densities in the former and latter areas were equal to $3 \times 10^{20}$ and $10^{20}$ cm$^{-3}$, respectively.

It is pertinent to note that the nonlinear processes under consideration may, in the general case, also give rise to higher-order harmonics of different intensity. In this case, the nice consistency of experimental and simulation data obtained with the inclusion of only the second harmonic allows the following conclusion: the conditions required for the development of higher-order harmonics with intensities comparable to the second harmonic intensity are not formed in the plasma under the pulsed laser intensities involved.

The efficiency of second harmonic generation should become lower with a decrease in laser intensity. This must entail the disappearance of plasma satellites shifted by the double frequency and the emergence of satellites shifted by the fundamental laser frequency. This statement was borne out by an experiment performed on the JLITE-X facility. In this case, the laser flux was almost an order of magnitude lower and the recorded spectra (Fig. 3) were nicely described by the simulations performed with the inclusion of only the monochromatic field.

Therefore, in this work concerned with the interaction of high-intensity laser radiation with cluster targets we showed that the formation of X-ray emission spectra may be affected by the bichromatic oscillating electric field related to the generation of the second harmonic of laser radiation. Investigating the subtle features of the spectral line profiles of multiply charged ions permits determining the structure of this field and, in particular, estimating the efficiency of second harmonic generation in laser-cluster interactions.

***Acknowledgements.*** This work was performed in the JIHT, RAS, with financial support from the Russian Science Foundation (Grant No. 14-50-00124).

## References

1. Negus C.R., Peacock N.J. *J. Phys. D*, **12**, 91 (1979).
2. Burkhalter P., Davis J., Rauch J., et al. *J. Appl. Phys.*, **50**, 705 (1979).
3. Boiko V.A., Vinogradov A.V., Pikuz S.A., Skobelev I.Yu., Faenov A.Ya. *Itogi Nauki i Tekhniki. Ser. Radiotekhnika* (Moscow: VINITI, 1980), Vol. 27.
4. Pikuz S.A., Sinars D.B., Shelkovenko T.A., et al. *Phys. Rev. Lett.*, **89**, 035003 (2002).
5. Kinkhabwala A., Sako M., Behar E., et al. *Astrophys. J.*, **575**, 732 (2002).
6. Abdallah J. Jr, Csanak G., Fukuda Y., et al. *Phys. Rev. A.*, **68**, 063201 (2003).
7. Vrielmann S., Ness J.-U., Schmitt J.H.M.M. *Astronom. Astrophys.*, **439**, 287 (2005).
8. Robrade J., Schmitt J.H.M.M. *Astronom. Astrophys.*, **449**, 737 (2006).
9. Nazé Y., Rauw G. *Astronom. Astrophys.*, **490**, 801 (2008).
10. Ryazantsev S.N., Skobelev I.Yu., Faenov A.Ya., et al. *Pis'ma Zh. Eksp. Teor. Fiz.*, **102**, 817 (2015).
11. Faenov A.Ya., Colgan J., Hansen S.B., et al. *Scientif. Rep.*, **5**, 13436 (2015).
12. Nishiuchi M., Sakaki H., Esirkepov T.Zh., et al. *Phys. Plasmas*, **22**, 033107 (2015).
13. Belyaev V.S., Vinogradov V.I., Kurilov A.S., et al. *Pis'ma Zh. Eksp. Teor. Fiz.*, **78**, 1216 (2003).
14. Gavrilenko V.P., Belyaev V.S., Kurilov A.S., et al. *J. Phys. A*, **39**, 4353 (2006).
15. Oks E., Dalimier E., Faenov A.Ya., et al. *J. Phys. B*, **47**, 221001 (2014).
16. Oks E., Dalimier E., Faenov A.Ya., et al. *Opt. Express*, **23**, 31991 (2015).
17. Fukuda Y., Faenov A.Ya., Pikuz T., et al. *Appl. Phys. Lett.*, **92**, 121110 (2008).
18. Kiriyama H., Mori M., Nakai Y., et al. *Opt. Lett.*, **35**, 1497 (2010).
19. Colgan J., Abdallah J. Jr, Faenov A.Ya., et al. *High Energy Density Phys.*, **7**, 77 (2011).
20. Boldarev A.S., Gasilov V.A., Faenov A.Ya., et al. *Rev. Sci. Instrum.*, **77**, 083112 (2006).
21. Jinno S., Fukuda Y., Sakaki H., et al. *Appl. Phys. Lett.*, **102**, 164103 (2013).
22. Faenov A.Ya., Pikuz S.A., Erko A.I., et al. *Phys. Scripta*, **50**, 333 (1994).
23. Blasco F., Stenz C., Salin F., et al. *Rev. Sci. Instrum.*, **72**, 1956 (2001).
24. Lavrinenko Ya.S., Morozov I.V., Pikuz S.A., Skobelev I.Yu. *J. Phys. CS*, **653**, 012027 (2015).